行政院國家科學委員會專題研究計畫 成果報告

東海長期觀測與研究(Ⅲ)：河川與大氣輸送物質對生物地球化學作用之影響--二氧化碳及海氣交換
研究成果報告(精簡版)

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行政院國家科學委員會補助專題研究計畫進度報告

二氧化碳及海氣交換（1/3）

Distribution and Air-Sea Exchange Fluxes of CO_2 in the East China Sea (1/3)

計畫類別： 整合型計畫

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I. 中文摘要：

本計畫是「東海長期觀測與研究（三）」的一項子計畫，為期三年。本計畫的目的在探討東海之二氧化碳的海氣交換循環及通量，利用大面積密集觀測及重點實驗整合的手法，來瞭解二氧化碳在大陸、海洋交界邊緣海域的循環及海氣交換的過程作用，並整個東海陸棚在調節大氣人為二氧化碳所佔之角色。本研究著重在立即船測二氧化碳分壓空間分布資料，計算二氧化碳海氣交換之通量，並藉以瞭解長江逕流及黑潮湧升流以及大氣外力對東海生地化的季節變化、空間變異等之影響，然後評估東海控制二氧化碳通量的主要機制。長期累積的觀測資料有助我們發展一個生地化及物理水文結合的模式來模擬及預測二氧化碳在東海海域的搬運及循環模式。最後，可成功地將研究結果應用到其他相似的邊緣海域，並提供政府單位有個有效及準確的環境決策方針。

關鍵詞：東海，二氧化碳，生地化循環，海氣交換通量，大陸/海洋邊緣

This is a three-year project, which is a part of the integrated research, “Long-term observation and Research of the East China Sea (III)” (LORECS). The main purpose of this project is to investigate the cycling and air-sea exchange of carbon dioxide (CO_2) and related controlling processes in the East China Sea (ECS) and further to examine the role of East China Sea margins in the global carbon cycling. We are planning a comprehensive examination of CO_2 in the coast margins using a combining experimental design (i.e, field investigations with a theoretic modeling). The main effort of observational study is devoted to carry out the underway fCO_2 measurements in the ECS. The results will be used to derive the air-sea exchange fluxes of CO_2, to understand the carbon dynamics of the East China Sea and further to evaluate the controlling mechanisms and impacts of Changjiang runoff and Kuroshio upwelling and atmospheric forcings on the CO_2 biogeochemistry of the ECS. Over the long-term observation, we expect to incorporate these multi-media data into a coupled physical-biogeochemical model for the ECS. The results will finally allow to be applicable to other similar coastal environments and to be further served as a decision tool for future environmental policy.

Key words: East China Sea, carbon dioxide, biogeochemistry, air-sea exchange flux, continental/ocean margins

II. Project description and scope

Global warming and climate change associated with increasing concentration of anthropogenic CO_2 in atmosphere is seriously and continuously concerned over the few decades (IPCC, 2001, Houghton et al, 2001). Since 1950's, many high-quality observation works and the analysis of air trapped in ice cores have revealed and well constrained the current and historical inventories and cycles of atmospheric CO_2 (Neftel et al., 1985; Indermühle et al., 1999; Keeling and Whorf, 2004). The large basin-scale carbon observation in the ocean was also implemented since 1990's WOCE. The goal of the global ocean carbon observation is to improve the understanding and prediction of the climate system by developing a network of sustained ocean observations. Under the conceptual framework of International Ocean Carbon Coordination Project held in Paris in 2003, ocean carbon community endeavored to coordinate the carbon observation activities, at an international levels, such as repeat hydrographic section programs like 1990's WOCE implements and the growing network of near-surface underway pCO_2 measurements together. The ultimate goal is to generate the high quality and resolution data for ocean carbon modeling and research activities associated with climate change. Above most observations are focusing on the open ocean carbon instead of the ocean/continental margins. That is, there is a critical gap occurred in the global carbon observation network concerning the important role of continental margins in the global oceanic carbon cycle (Walsh et al., 1981; Walsh, 1999; Tsunogai and Watanabe, 1999; Rabouille et al., 2001).

The evaluation on the role of continental margins in the ocean carbon cycle is quite limited with respect to the quantification and dynamics of fluxes (Chen and Wang, 1999; Tsunogai and Watanabe, 1999; Liu et al., 2000; Chen, 2003, Thomas et al., 2004). The continental margins are major receiving compartments for natural and anthropogenic derived inorganic/organic carbon pollutant from riverborne/watershed and atmospheric inputs (Martin and Windom, 1991). In addition, the waters interact strongly and in complex ways with the land, atmosphere, continental shelves and slopes, and the open ocean. The specific rates of productivity, biogeochemical cycling and organic/inorganic matter sequestration are higher in coastal margins than those in the open ocean. The high organic matter deposition to the sediments and interactions at the sediment-water interface raises the importance of sedimentary chemical redox reactions with implications for the global carbon, nitrogen, phosphorus and iron cycles. Thus, increased knowledge and understanding concerning the biogeochemistry and flux dynamics of carbon and its anthropogenic impact on ocean carbon cycle in biologically productive marginal regions are critically needed. More importantly, the current lack of knowledge and understanding of air-sea CO_2 exchange processes occurring at the continental margins has left them largely ignored in most of the previous global assessments of the oceanic carbon cycle. Recently, contrasting and conflicting results have been reported regarding whether ocean margins are a source or a sink of atmospheric CO_2. This stems from a long-standing debate as to whether coastal oceans are heterotrophic or autotrophic biological systems.

A three-year proposal for near-surface underway pCO_2 observation study related to gas-exchange flux and controlling factors affecting the carbon cycle in the productive East China Sea (ECS) was thus proposed. The ECS provides an ideal coastal laboratory in which to examine the specific processes (e.g., solubility and

biological pump and continental shelf pump) and important interfacial exchanges with the coastal zone (e.g., rivers and estuaries, air-sea exchange etc.) of carbon and its subsequent behavior and fate. The related conceptual model to integrate continental margin carbon fluxes and further assess the anthropogenic influence (e.g., waste waters from river discharge, atmospheric CO_2 to the surface ocean etc.) on the ocean carbon cycle may be assessed afterwards. Finally, the outcome of the carbon research makes helping to quantify critical gaps in the understanding of carbon in the coastal environment and recognition of the significance of continental margins in processing carbon and controlling of anthropogenic CO_2.

III. Preliminary results

As part of our general interests and efforts concerned with the global carbon cycling with climate change, we have been investigating the cycling and air-sea exchange of CO_2 over the ocean margins around Taiwan. A significant part of this work has been conducted within the SEATS (SouthEast Asian Time-series Study) program supported by the NSC since 1998. The SEATS is a multidisciplinary, coordinated research project to examine the role of near tropic oligotrophic oceans in the global change and investigate the processes dominating the fate and behavior of carbon and climate-relevant elements. Interesting findings about the dynamics and air-exchange flux of carbon associated with biogeochemical response to external forcings in the euphotic zone of the ocean were obtained over the past years. Part of carbon research activities was carried out in East China Sea shelf by the LORECS integrated program. Some interesting results associated with dynamics and air-sea exchange of CO_2 are still generated and fairly presented in professional meetings. The above two investigated areas are ocean margins but have different environmental settings so that the occurrence of carbon with respect to the distribution and air-sea exchange flux vary each other. Summary of both results in the SCS and ECS are listed in the following.

A. Air-sea CO_2 exchange at the SEATS station in northern South China Sea (Tseng et al., 2007, Deep-Sea Research II)

Carbonate chemistry has been studied at the SEATS station on bimonthly to quarterly cruises since September 1999. More recently underway PCO_2 measurements have been conducted on the time-series cruises. The surface fCO_2, calculated from the TCO_2, TA, phosphate and silicate data for samples collected during our cruises, varies seasonally. The monthly pattern of fCO_2 concentration gradient (i.e., $fCO_2 = fCO_{2\ water} - fCO_{2\ air}$) which represents a driving potential for CO_2 gas transfer across water- air interface show that positive fCO_2 occurs from March to October, signifying a loss of fCO_2 from the SCS to the atmosphere, and uptake of atmospheric CO_2 during the rest of the year. The air-sea exchange fluxes of fCO_2, F, between the atmosphere and surface waters of the SCS are estimated using the following equation: $F = K (fCO_{2\ water} - fCO_{2\ air})$, where K (gas transfer coefficient) $= \kappa L$ (κ: transfer velocity; L: gas solubility), fCO_{2w} and fCO_{2a} are respectively the fugacity of CO_2 in the surface waters and overlying air. The κ is derived using the following empirical relationship developed by Wnninkkhof (1992). Solubility of CO_2 can be estimated from the empirical equation (Weiss, 1974) by sample temperature and salinity. The monthly wind speed data at 10 m height at SEATS are computed from the ECMWF database of 1985-2003.

Preliminary results of a 4-year period work show the air-sea flux of CO_2 vary greatly in different seasons (Fig. 1). The general pattern shows the average upward flux of 1.1±0.2 mol m^{-2} y^{-1} in summer and average downward flux of -1.8±0.4 mol m^{-2} y^{-1} in winter. The seasonal changes in fCO_2 may be accounted for mostly by the seasonal changes in water temperature (~6 °C). However, the associated air-sea exchange flux is also controlled by wind speed, which is much stronger in winter. Overall speaking, the air-sea exchange of CO_2 at SEATS station is nearly at balance, but the stronger winter monsoon, under which observations are scarce, may make the net transfer a very weak sink (around -0.01 mol C m^{-2} y^{-1}). This averaged value is quite less than that uptake rates reported in the earlier by Takahashi et al. (2002) in the SCS region (ca. ~0 to ~0.4 mol C m^{-2} y^{-1}) and by Chou et al. (2005) (ca. ~0.1 to ~0.2 mol C m^{-2} y^{-1}).

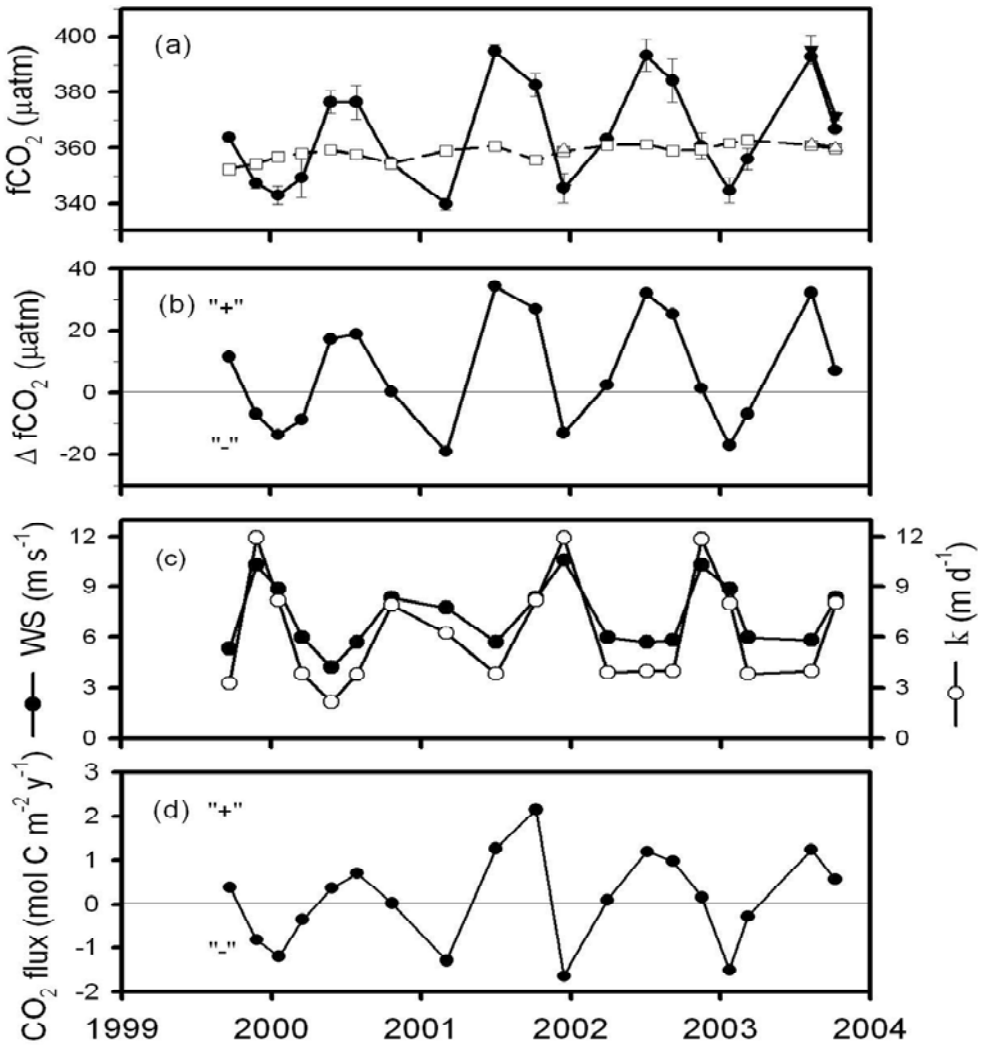

Figure 1 Variations in (a) the average fCO_{2w} (• –calculated; ▼– underway) and fCO_{2a} (□ – estimated; △ – underway) in the mixed layer, (b) the average ΔfCO_2 in the mixed layer, (c) the monthly average climatological wind speed (•) and gas transfer velocity (k) (o), and (d) CO_2 flux in different sampling months in the year ("+" : sea to air; "–" : air to sea) at the SEATS station between

September, 1999 and October, 2003.

B. Air-sea CO_2 exchange in East China Sea (Tseng et al., unpublished)

The fCO_2 underway survey in the East China Sea was carried out during cruise of OR1-836 in July 2007 (Fig. 2). Sea surface pCO_2 distribution in July 2007 showed significant spatial and temporal variation, ranging from 172 to 554 μatm with an average of 349±71 (n=1596). Low pCO_2 levels were found in low salinity coastal waters off Mainland China. High nutrient loadings from Changjiang and Mingjiang Rivers appeared to be the most important factor contributing to the drawdown of the coastal seawater pCO_2 values biogeochemically. Air pCO2 levels remained fairly constant with an average of 372±2 μatm. The pCO_2 concentration gradient (i.e. delta $pCO_2 = pCO_{2\ water} - pCO_{2\ air}$) ranged from –204 μatm to 178 μatm with an average of –23 ± 71 μatm. Most of samples were undersaturated relative to the atmosphere during July, 2007.

In general >50% of China coast regions are CO_2 sink especially near river mouths. Strong CO_2 sink probably was due to riverine source of which nutrient was high and pCO_2 was low, and high nutrient water was brought up from subsurface water. These high nutrient waters induce strong biological activity (high Chlorophyll a) and thus reduce CO_2 in the euphotic zone. Phytoplankton bloom events were usually found during the cruise especially near river mouths. However, the correlation of high chlorophyll a versus low pCO_2 seems to be not the case for locations near the upwelling site where chlorophyll-a was high but pCO_2 was high too. This phenomenon indicates that physical factor such as CO_2 supplying rate due to upwelling event and the biological uptake rate of CO_2 are both important factors to determine pCO_2 value in the surface.

Based on Winninkhof's (1992) empirical equation, the CO_2 invasion flux F can be estimated as the product of the gas transfer velocity (k), the solubility (α), and the difference of CO_2 fugacity between air and water: $F = k\alpha(fCO_{2\ water} - fCO_{2\ air})$.Taking the mean wind speed of 7±1 m s^{-1} in June in the ECS computed from the Taiwan CMB database of 1999-2007, the equation gives air-sea CO_2 flux of 1±2 mol m^{-2} y^{-1}. Overall, there will be an annual CO_2 uptake of 0.01 Gt C by the ECS, if ECS area is taken to be 1.25 x 10^{12} m^2. The result is similar to the value of 0.03 Gt C/yr estimated by Peng et al. (1999) based on the data collected in the spring. It further indicates that the shelf regions could be significant CO_2 sinks. To have a better understanding and quantification of the ECS as a sink of CO_2, more investigations are needed for all other seasons of the year to obtain a representative estimate.

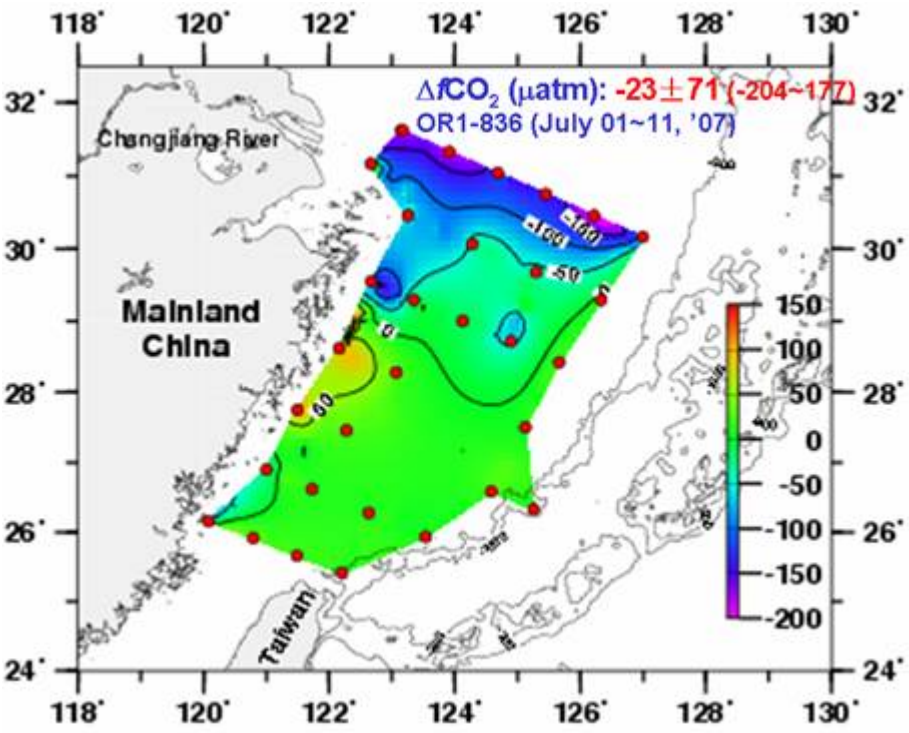

Fig.2 Distribution contour of delta fCO2 (i.e. $fCO_{2\ water}$- $fCO_{2\ air}$) in the ECS

List of publications associated with carbon cycling in coastal margins

1. Wong, G. T. F., T.-L. Ku, M. Mulholland, C.-M. Tseng, D.-P. Wang (2007) The SouthEast Asian Time-series Study (SEATS) and the biogeochemistry of the South China Sea – An Overview, *Deep-Sea Research II*, 54/14-15: 1434-1437. (SCI:1.278, NSC 94-2119-M-002-027)

2. Tseng C.-M., G.T.F. Wong, W.-C. Chou, B.-S. Lee, D.D. Sheu, K.-K. Liu (2007) Temporal Variations in the carbonate system in the upper layer at the SEATS station. *Deep-Sea Research II*, 54/14-15: 1448-1468. (SCI:1.278, NSC 94-2119-M-002-027; NSC 94-2611-M-002-019)

3. Liu, K.-K., S.-J. Kao, H.-C. Hu, W.-C. Chou, G.-W. Hung, C.-M. Tseng (2007) Carbon isotopic compositions of suspended and sinking particulate organic matter in the northern South China Sea from production to deposition. *Deep-Sea Research II*, 54/14-15: 1504-1527. (SCI:1.278, NSC 94-2119-M-002-027)

4. Liu, K.-K., Y.-J. Chen, C.-M. Tseng, I.-I. Lin, H.-B. Liu, A. Snidvongs (2007) The significance of phytoplankton photo-adaptation and benthic-pelagic coupling to primary production in the South China Sea: Observations and numerical investigations, *Deep-Sea Research II*, 54/14-15: 1546-1574. (SCI:1.278, NSC 94-2119-M-002-027)

5. Wong, G. T. F., C.-M. Tseng, L.-S. Wen, S.-W Chung (2007) Nutrient dynamics

and nitrate anomaly at the SEATS station. *Deep-Sea Research II*, 54/14-15: 1528-1545. (SCI:1.278, NSC 94-2119-M-002-027)

6. Chou, W.-C., D. D. Sheu, B. S. Lee, C.-M. Tseng, C. T. A. Chen, S. L. Wang, G. T. F. Wong (2007) Depth distributions of alkalinity, TCO_2 and $d^{13}C_{TCO2}$ at the SEATS time-series site, northern South China Sea: controlling processes and anthropogenic CO_2 influence. *Deep-Sea Research II*, 54/14-15: 1469-1485. (SCI:1.278, NSC 94-2119-M-002-027)

7. Liu, H., J. Chang, C.-M. Tseng, L.-S. Wen, K. K. Liu (2007) Seasonal variability of picoplankton in the northern South China Sea at the SEATS station. *Deep-Sea Research II*, 54/14-15: 1602-1616. (SCI:1.278, NSC 94-2119-M-002-027)

8. Liu, K.-K., C.-M. Tseng, C.-R. Wu, I-I Lin (2007) Biogeochemical fluxes of the South China Sea. In: Liu, K.-K., Atkinson, L., Quiñones, R., Talaue-McManus, L. (Eds.) Carbon and nutrient fluxes in continental margins: a global synthesis. IGBP Book Series, Springer. (Book chapter, NSC 93-2119-M-002-032)

9. W.C. Chou, Y.L. Lee, Y.Y. Shih, D.D. Sheu, C.A. Han, C.L. Cho, C.-M. Tseng, Y.J. Yang (2006) Estimated net community production during the summer time at the SEATS time-series study site, northern South China Sea: implications for nitrogen fixation, *Geophysical Research Letters*, 33, L22610,doi: 10.1029/2005GL025365. (SCI:2.491, NSC 94-2119-M-002-027)

10. Chen C-C, K-P Chiang, G-C Gong, F-K Shiah, C.-M. Tseng, K-K Liu (2006) The importance of planktonic community respiration on the carbon balance of the East China Sea in summer, *Global Biogeochemical cycles*, 20, GB4001, doi:10.1029/2005GB002647 (SCI:3.373, NSC 95-2611-M-002-003)

11. Wen-Chen Chou, David D., Sheu, C.T. Arthur Chen, S. L. Wang and C.-M. Tseng (2005) Seasonal variability of carbon chemistry at the SEATS time-series site, northern South China Sea between 2002 and 2003. *Terrestrial, Atmospheric and Oceanic Sciences*, 16, 445-465. (SCI:0.965; NSC 93-2119-M-002-032)

12. Tseng C.-M., George T.F. Wong, I. I. Lin, Chou-Long Wu, KK Liu (2005) A unique seasonal pattern in phytoplankton biomass in low-latitude waters in the South China Sea, *Geophysical Research Letters*, 32, L08608, doi:10.1029/2004GL022111. (SCI:2.491; NSC 93-2119-M-002-032)

Published abstracts of papers presented at professional meetings

1. Chun-Mao Tseng, B.-S. Lee, W.-C. Chou, D.D. Sheu, G.T.F. Wong, K.-K. Liu (2004) Temporal Variability of Upper-layer Oceanic CO_2 Species at SEATS, 2004 West Pacific

Geosciences Meeting (WPGM). Honolulu, Hawaii, August 16~20.

2. Y.J. Chen, Chun-Mao Tseng, S.J. Kao, K.K. Liu (2004), Particulate organic carbon and nitrogen at SEATS station: observations and numerical modeling, 2004 West Pacific Geosciences Meeting (WPGM), Honolulu, Hawaii, August 16~20.

3. Chun-Mao Tseng, B.-S. Lee, W.-C. Chou, D.D. Sheu, G.T.F. Wong, K.-K. Liu (2005) Temporal Variability of Upper-layer Oceanic CO_2 Species at the SEATS, American Society of Limnology and Oceanography (ASLO) 2005 Aquatic Sciences Meeting. Feb 20-25, 2005, Salt Lake City, Utah, USA..

4. Chun-Mao Tseng, B.-S. Lee, W.-C. Chou, D.D. Sheu, G.T.F. Wong, K.-K. Liu (2005) Temporal Variability of Upper-layer Oceanic CO_2 Species at the SEATS, AGU Chapman Conference on the role of marine organic carbon and calcite fluxes in driving global climate change, past and future, July 24-27, 2005, WHOI, Woods Hole, USA.

5. Chun-Mao Tseng, G.-C. Gong, K.-K. Liu (2006) Distribution and air-sea exchange fluxes of CO_2 in the East China Sea, 2006 Ocean Science Meeting, AGU Honolulu, Hawaii, February 20~24.