

# Methane distributions and sea-to-air fluxes in the South China Sea and the West Philippines Sea

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## Introduction

Methane ( $\text{CH}_4$ ), an atmospheric trace gas, has a global warming potential in a 100-year time frame ( $\text{GWP}_{100}$ ) that is 34 times that of carbon dioxide ( $\text{CO}_2$ ).  $\text{CH}_4$  is the most abundant hydrocarbon in the atmosphere and plays an important role in atmospheric chemistry. The South China Sea (SCS) is the largest marginal sea in the world and is connected with the West Philippines Sea (WPS) via the Luzon Strait, which is around 2200 m in depth and is the deepest sill that connects the SCS with any adjacent body of water. This work elucidates the spatial and vertical distributions of  $\text{CH}_4$  in the water column of the SCS and WPS, as well as its sea-to-air fluxes, and further discusses differences between these seas and the factors that influence those distribution and fluxes.

## Materials and Methods

Samples from the SCS were taken during six cruises on board R/V Ocean Researchers I and III (Fig. 1a); samples from the WPS were collected during three cruises (Fig. 1b). Fluxes of  $\text{CH}_4$  across the air-water interface were estimated using  $F=k(C_w - \alpha C_a)$  where  $k$  ( $\text{cm h}^{-1}$ ) is the gas exchange coefficient, as described by Wanninkhof (2014). Wind speeds at 10 m above from the surface were obtained from the NASA QuikSCAT satellite platform. The seawater fluxes from the SCS to the WPS were calculated as described by Chen *et al.* (2001).

## Results and Discussion

- The surface  $\text{CH}_4$  concentration that was measured during the wet seasons in the WPS was  $3.0 \pm 1.2 \text{ nM}$  (saturation 155%) and in the SCS was  $4.5 \pm 3.6 \text{ nM}$  (saturation 230%); the sea-to-air  $\text{CH}_4$  fluxes in wet season in the SCS was  $8.6 \pm 12.7 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$  and in the WPS was  $4.9 \pm 4.9 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$  (Table 1).
- The maximum  $\text{CH}_4$  and chlorophyll  $a$  concentrations were found in the subsurface waters (Figs. 2a and 2b) in the WPS. Although the formation of  $\text{CH}_4$  in the surface water layer may relate to the biological activity, no significant correlation between  $\text{CH}_4$  and chlorophyll  $a$  concentrations in the surface 200 m layer was identified herein (Fig. 4). This result not only shows that the formation of  $\text{CH}_4$  may not be directly associated with the photosynthetic process or phytoplankton biomass but it also reveals that other processes, such as the physical mixing of different water masses, or  $\text{CH}_4$  inputs from seeps or sediments, may affect the distribution of  $\text{CH}_4$  in the WPS.
- Since little biogenic  $\text{CH}_4$  is produced outside of the euphotic zone,  $\text{CH}_4$  concentrations generally decline to undersaturated levels with increasing depth beyond the euphotic zone and remain constant below 1,000 m (Figs. 2a and 3b). Some high  $\text{CH}_4$  concentrations were measured in the mid-depths of the SCS (Fig. 3b).
- According to Chen *et al.* (2001), in the wet season, surface and intermediate waters are net-exported from the SCS to the WPS but deep water is net-imported. The net export of surface and intermediate waters carry  $1.5 \times 10^6 \text{ mol d}^{-1}$  and  $0.53 \times 10^6 \text{ mol d}^{-1} \text{ CH}_4$  from the SCS to the WPS in the wet season while the deep water from the WPS imports  $0.15 \times 10^6 \text{ mol d}^{-1} \text{ CH}_4$  to the SCS (Table 2).

## Conclusions

The SCS influences the surface and intermediate waters in the WPS. This study has established that the SCS emits  $\text{CH}_4$  to the atmosphere and also exports  $\text{CH}_4$  to the WPS, and the sea-to-air flux is larger than the horizontal flux in the wet season. However, the  $\text{CH}_4$  distribution and flux during the dry season could not be determined because data are not available.

Table 1. Surface  $\text{CH}_4$  concentrations and sea-to-air fluxes in the SCS and the WPS in the wet season

	Latitude (°N)	Longitude (°E)	Sal.	Average Surface $\text{CH}_4$ Concentration (nM)	Wind Speed (m/s)	Sea to Air Flux ( $\mu\text{mol m}^{-2} \text{ d}^{-1}$ )	Surveying Cruises/Months
Domain SCS	A	20-25	117-120	$33.86 \pm 0.34$	$4.5 \pm 2.3 \text{ (n=31)}$	$7.2 \pm 1.8$	$9.9 \pm 8.7$ ORIII-896 (Aug. 2003) ORIII-983 (Jul. 2004)
	B	17-23	112-117	$33.17 \pm 1.08$	$5.2 \pm 2.1 \text{ (n=42)}$	$7.0 \pm 1.3$	$12.0 \pm 7.4$ ORI-695 (Sep. 2003) ORIII-1081 (Jul. 2005) ORI-802 (Jul. 2006)-Stn.SCS1
	C	3-12	104-113	$33.23 \pm 0.19$	$4.9 \pm 1.5 \text{ (n=19)}$	$6.2 \pm 1.0$	$8.3 \pm 4.1$ ORI-802 (Jul. 2006)
	D	4-20	110-120	$32.87 \pm 1.00$	$3.4 \pm 1.0 \text{ (n=30)}$	$6.3 \pm 0.9$	$4.3 \pm 3.9$ ORI-837 (Jul. 2007)
WPS	Avg.	3-25	104-120	$33.28 \pm 1.5$	$4.5 \pm 3.6 \text{ (n=122)}$	$6.3 \pm 0.8$	$8.6 \pm 12.7$ ORI-896 (Aug. 2003) ORI-695 (Sep. 2003) ORI-983 (Jul. 2004) ORI-1081 (Jul. 2005) ORI-802 (Jul. 2006) ORI-837 (Jul. 2007)
	Avg. Continental Shelf region (depth<500m)		33.29±0.9	5.1±2.1 (n=72)		$11.0 \pm 7.4$	ORI-725 (Aug. 2004) ORI-1149 (May 2006)
	Avg. Deep Sea (depth>500m)		33.54±0.7	3.8±1.6 (n=50)		$6.1 \pm 6.0$	ORI-837 (Jul. 2007)-Stn.A-D;1-5, unpublished data from C. T. A. Chen

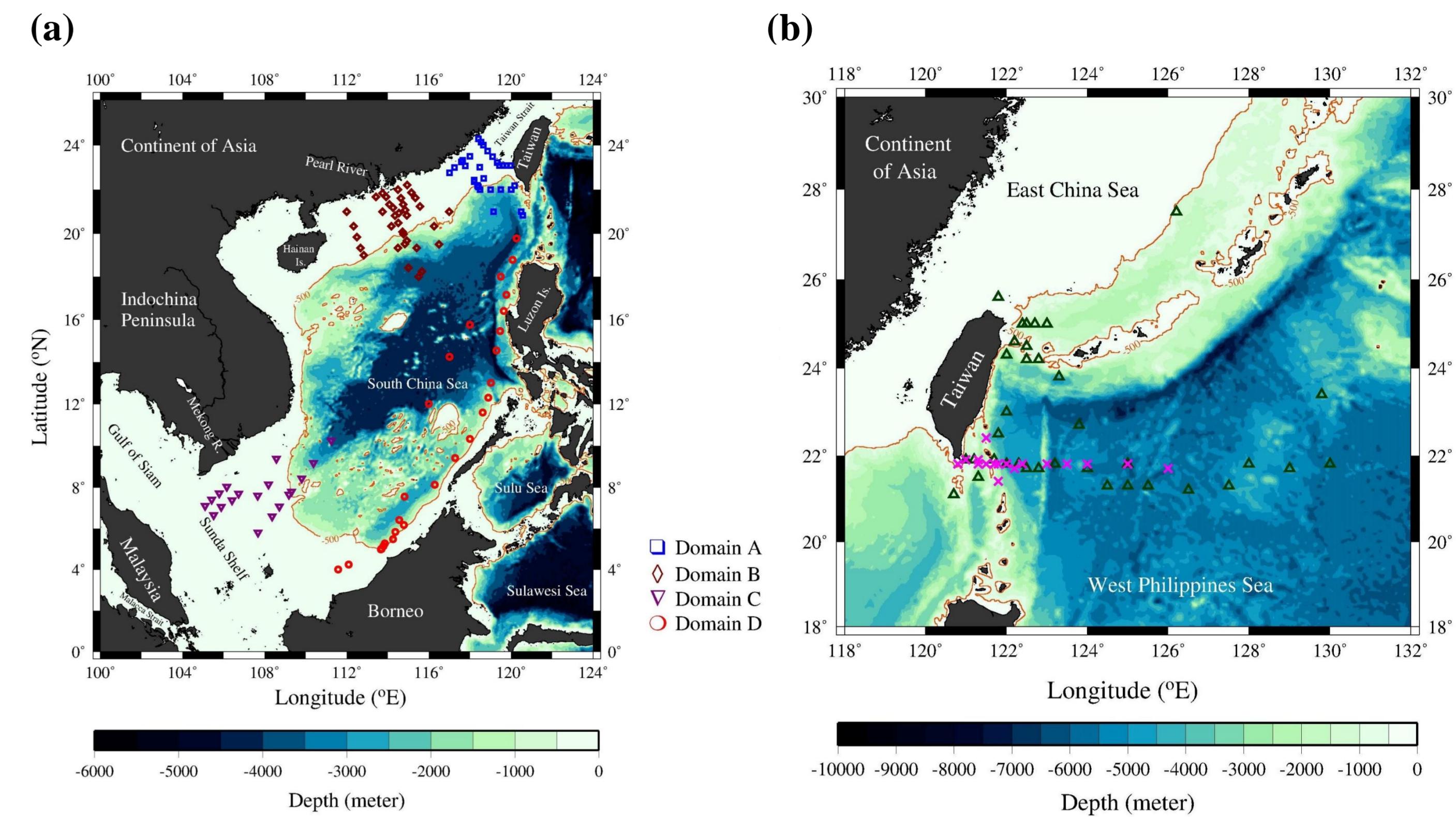


Fig. 1. Study area and station locations:

(a) South China Sea stations. □: Domain A, ORIII-896 (Aug. 2003) and ORIII-983 (Jul. 2004); ◇: Domain B, ORI-695 (Sep. 2003), ORIII-1081 (Jul. 2005) and ORI-802 (Jul. 2006)-Stn.SCS1; ▽: Domain C, ORI-802 (Jul. 2006); ○: Domain D, ORI-837 (Jul. 2007);

(b) West Philippines Sea stations. △: surface sampling stations, ORI-725 (Aug. 2004), ORI-837 (Jul. 2007) -Stn.A-D;1-5 and unpublished data from C. T. A. Chen; ×: discrete sampling stations, ORI-725 (Aug. 2004) and ORIII-1149 (May 2006)

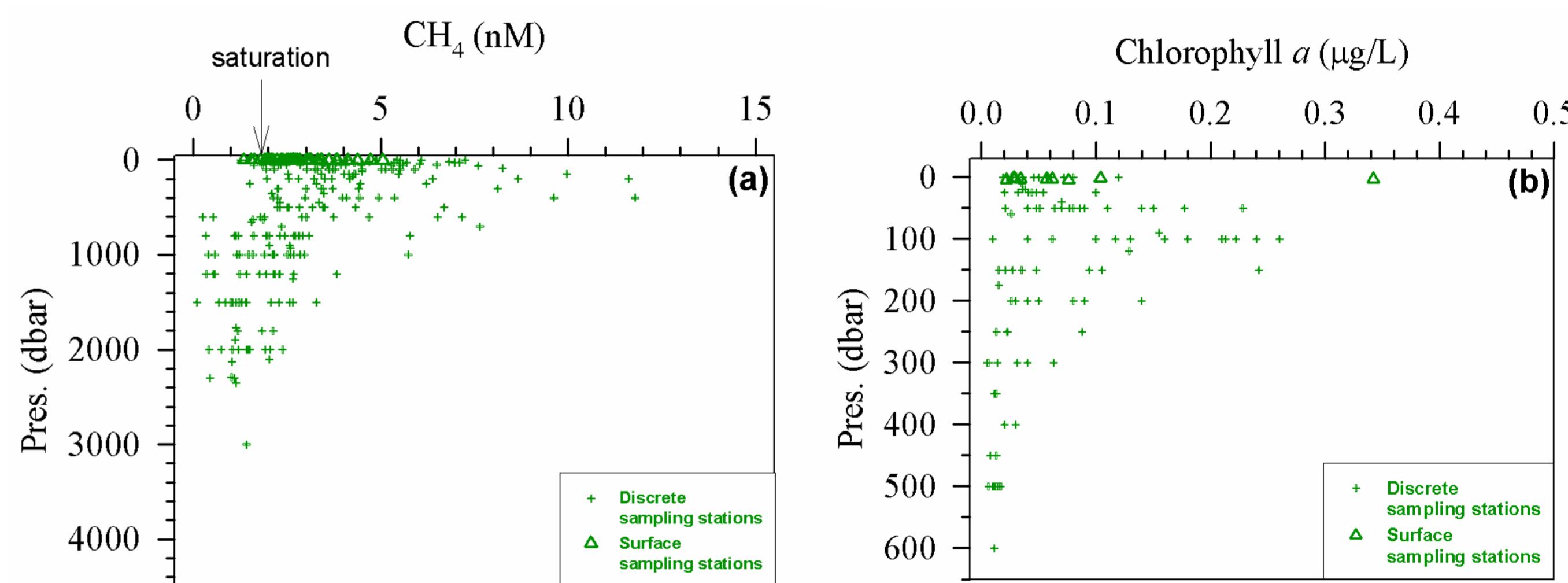


Fig. 2. Vertical distributions of (a)  $\text{CH}_4$  (nM) and (b) chlorophyll  $a$  ( $\mu\text{g L}^{-1}$ ) in the West Philippines Sea, obtained at 43 stations during three cruises from August 2004 to July 2007.

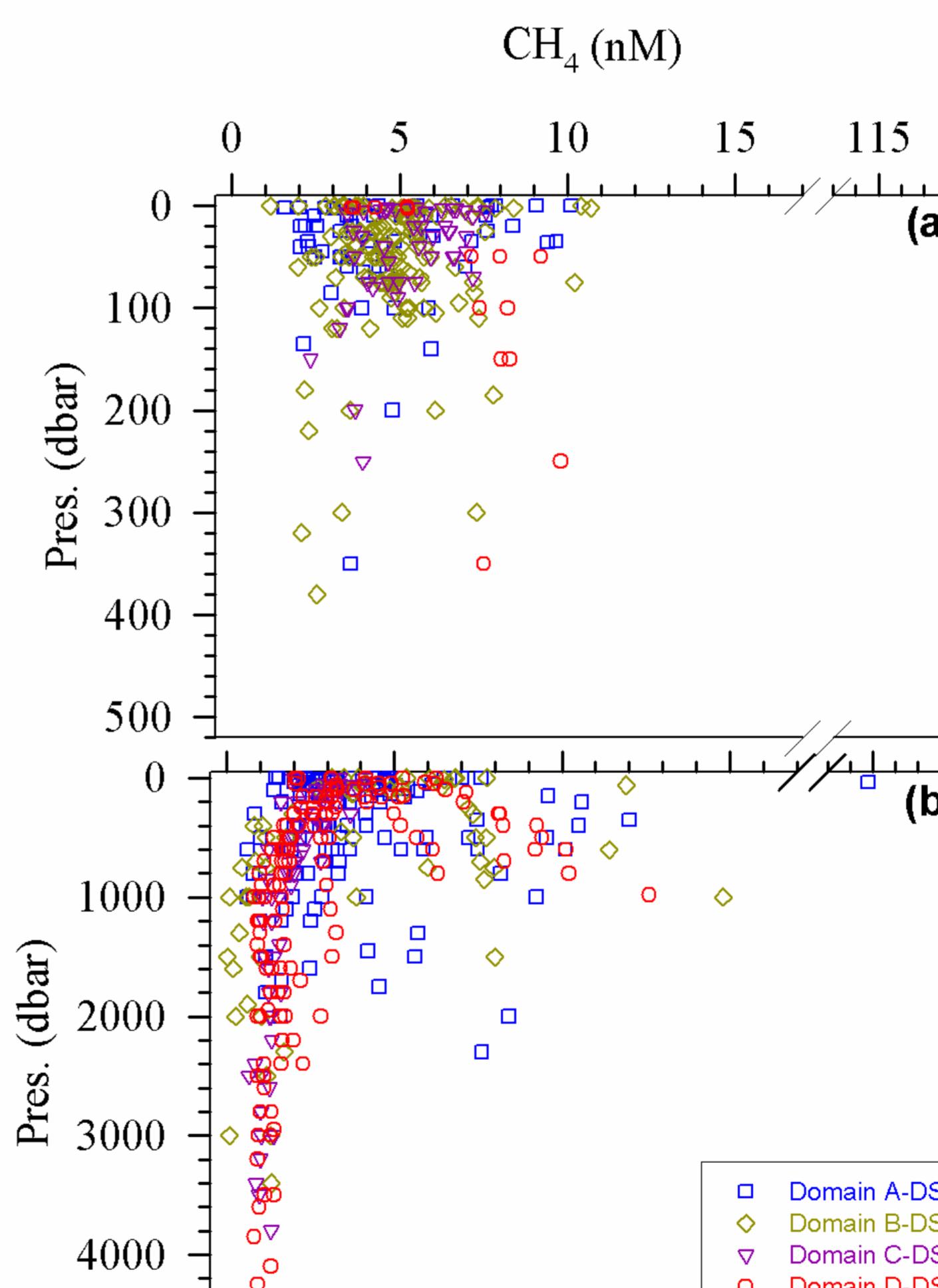


Fig. 3. Vertical distributions of  $\text{CH}_4$  (nM) (a) on the continental shelf and upper slope (depth<500m) and (b) in the deep sea (depth>500m) in the South China Sea, obtained at 122 stations during six cruises from August 2003 to July 2007.

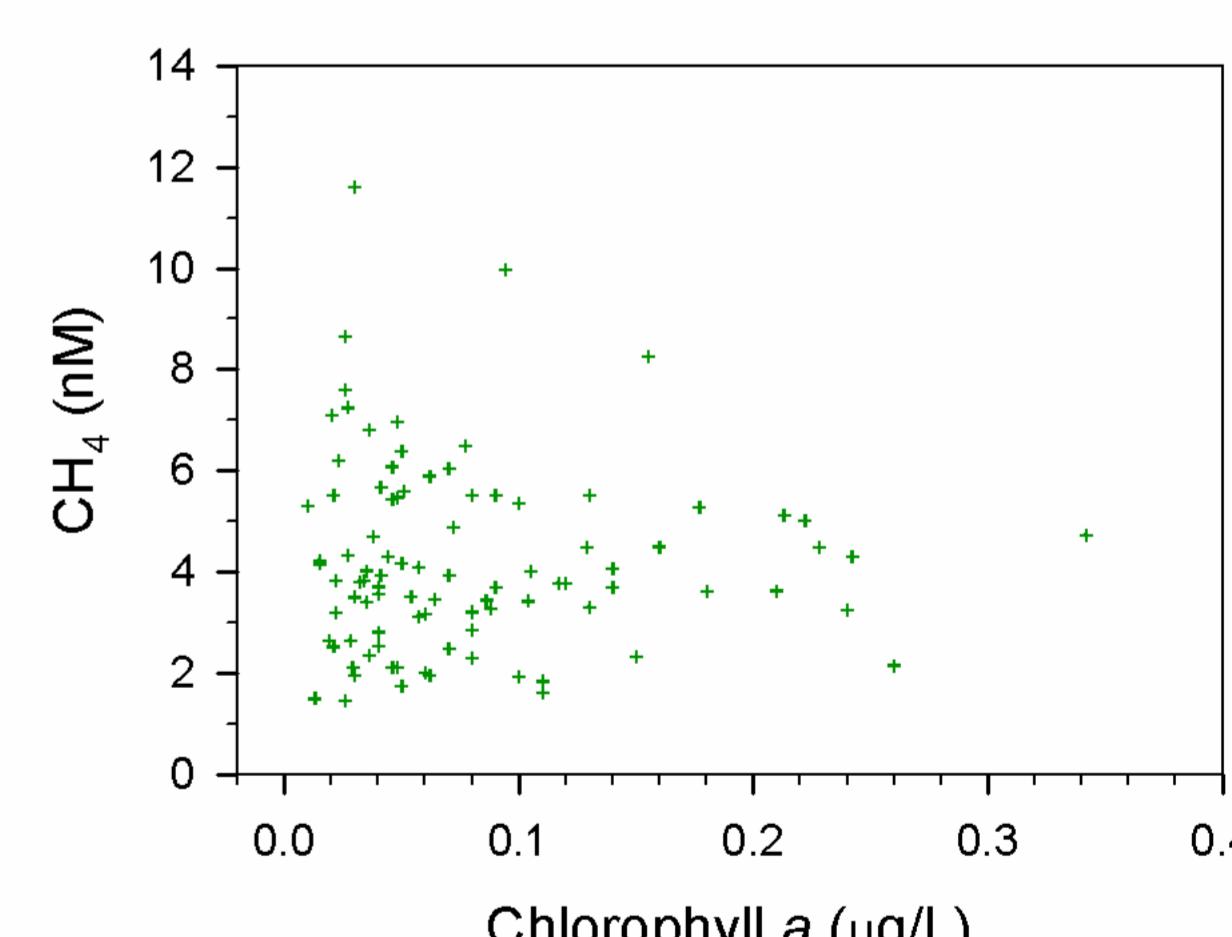


Fig. 4.  $\text{CH}_4$  (nM) versus chlorophyll  $a$  ( $\mu\text{g L}^{-1}$ ) in the surface 200 m layer in the West Philippines Sea, obtained at 43 stations during three cruises from August 2004 to July 2007.

Table 2.  $\text{CH}_4$  concentrations, water flux and  $\text{CH}_4$  flux of various water masses in the SCS and WPS

	SCS			WPS		
	Avg. Concentration nM	Water Flux $\times 10^6 \text{ t s}^{-1}$	$\text{CH}_4$ Flux $\times 10^6 \text{ mol d}^{-1}$	Avg. Concentration nM	Water Flux $\times 10^6 \text{ t s}^{-1}$	$\text{CH}_4$ Flux $\times 10^6 \text{ mol d}^{-1}$
Surface Water Layer (0-350m)	$4.8 \pm 5.7$	$-13.9 \pm 1.8$	$-5.78$	$3.9 \pm 1.9$	$12.8 \pm 1.1$	$4.28$
Intermediate water Layer (350-1350m)	$3.4 \pm 3.0$	$-1.8 \pm 0.4$	$-0.53$			
Deep Water Layer (1350-2200m)					$1.5 \pm 0.7$	$1.2 \pm 0.2$
						$0.15$

Positive and negative numbers represent inflow and outflow, respectively.

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