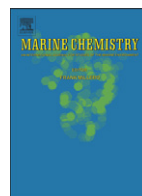




Contents lists available at ScienceDirect

Marine Chemistry

journal homepage: www.elsevier.com/locate/marchem

Enhancement of particulate organic carbon export flux induced by atmospheric forcing in the subtropical oligotrophic northwest Pacific Ocean

Chin-Chang Hung*, Gwo-Ching Gong, Wang-Chen Chung, Wei-Ting Kuo, Fu-Chen Lin

Institute of Marine Environmental Chemistry and Ecology, National Taiwan Ocean University, Taiwan

ARTICLE INFO

Article history:

Received 25 June 2008

Received in revised form 15 November 2008

Accepted 20 November 2008

Available online xxxx

Keywords:

POC

Export flux

Asian dust storm

Sediment trap

Pacific Ocean

ABSTRACT

The effects of extreme atmospheric forcing on the export flux of particulate organic carbon (POC) in the warm oligotrophic nitrogen-limited northwest Pacific Ocean were examined in 2007 during the spring Asian dust storm period. Several strong northeast monsoon events (maximum sustained wind speeds approaching 16.7 m s^{-1} , and gusts up to 19.0 m s^{-1}) accompanied by dust storms occurred during a 1-month period. The cold stormy events decreased surface water temperature and induced strong wind-driven vertical mixing of the water column, resulting in nutrient entrainment into the mixed layer from subsurface waters. As a result, the export flux of POC ranged from 49 to 98 (average value = $71 \pm 16 \text{ mg m}^{-2} \text{ day}^{-1}$), approximately 2–3 times greater than average values in other seasons. As dry and wet deposition of nitrogen attributable to Asian dust storm events does not account for the associated increases in POC stocks in this N-limited oligotrophic oceanic region, the enhancement of POC flux must have been caused by nutrient entrainment from subsurface waters because of the high winds accompanying the dust storm events.

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1. Introduction

Particulate organic carbon (POC) export fluxes in open ocean waters are generally low, typically averaging $<30 \text{ mg m}^{-2} \text{ day}^{-1}$ (Neuer et al., 2002). However, some natural events including dust storms, volcanic eruptions, typhoons, and hurricanes can induce significant material exchange between the ocean surface and the atmosphere. This can result in increased biological activity, such as increased primary production in oligotrophic regions and drawing down of CO_2 from the atmosphere (Lin et al., 2003; Uematsu et al., 2004; Gong et al., 2005; Herut et al., 2005). However, it is not known whether these natural extreme events transfer CO_2 from the atmosphere to the oceanic interior, because sampling is difficult under such weather conditions.

The subtropical northwest Pacific Ocean is an oligotrophic region, and, based on year-round observations (Gong et al., 1999), is nutrient-limited (nitrate below the detection limit of $0.1 \mu\text{M}$ in the upper 50 m) and has low chlorophyll *a* (Chl *a*) concentrations ($\leq 0.25 \text{ mg m}^{-3}$). Over six field investigations Gong et al. (1999) reported that phytoplankton biomass (based on Chl *a*) showed only small seasonal variations. During autumn and winter, when the northeast monsoon mixed the water column extensively, Chl *a* concentrations were distributed evenly in the euphotic zone and a subsurface Chl *a* maximum was not observed; in summer, high Chl *a* concentrations ($0.3\text{--}0.4 \text{ mg m}^{-3}$) were found close to the bottom of the euphotic zone (i.e., in the nitracline, approximately $100 \pm 10 \text{ m}$ deep) (Gong et al., 1999). The

main phytoplankton groups in the subtropical northwest Pacific Ocean are *Synechococcus* and *Prochlorococcus* (Chang et al., 1996; Chiang et al., 2002; Chang, unpublished data). The POC inventory (integrated to 125 m) is also low, and typically does not show marked seasonal variations ($3.0\text{--}3.6 \text{ g m}^{-2}$; Liu et al., 1995). In addition, phytoplankton productivity can also increase because of enhanced nutrient (i.e., N) deposition during dust storm events, but only in the surface layer to about 10 m depth (Gong et al., 2005). An explanation is that the subtropical oligotrophic northwest Pacific Ocean appears to be a nitrogen-limited system (Wu et al., 2003), and dissolved Fe concentrations ranging from $0.2\text{--}0.5 \text{ nM}$ (Wen et al., 2006) would be sufficient to support phytoplankton growth (Wells, 2003).

Asian dust storms, originating from the Gobi Desert in Mongolia and China, often occur in the spring. These storms blow eastward over Korea, Japan, and Taiwan, and can sometimes be so powerful that they spread dust across the Pacific Ocean as far as the United States (Uematsu et al., 1986; Sun et al., 2001). As increased phytoplankton growth, which is dependent on the availability of iron and other nutrients, coincides with these storms in the North Pacific, it may be that the dust fertilizes the ocean with bioavailable iron (DiTullio and Laws, 1991; Bishop et al., 2002; Honda, 2003; Yuan and Zhang, 2006). Dust storms not only carry dust, micro-nutrients, and pollutants, but also cause strong atmospheric–oceanic disturbances because of the extreme weather.

The influence of dust storms on the export flux of POC in the subtropical oligotrophic northwest Pacific Ocean is poorly understood because sea-based observations during storms are very difficult and dangerous as a result of the rough sea surface conditions. For this reason long-term field experiments are difficult to sustain. In recent

* Corresponding author.

E-mail address: cchung@ntou.edu.tw (C.-C. Hung).

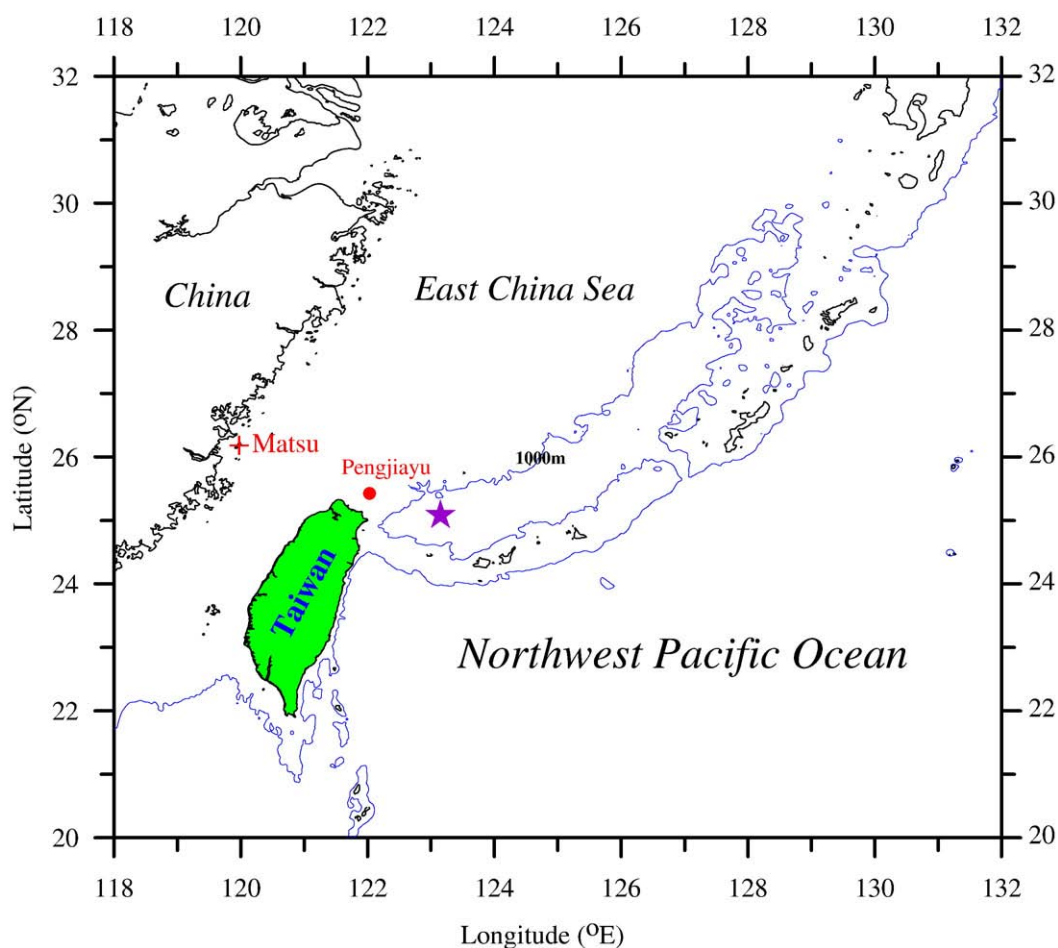


Fig. 1. Sampling location in the northwest Pacific Ocean. The samples were collected at Matsu Island; the wind speed was recorded at Pengjiayu Island; the star indicates the cruise area.

times the integrated LORECS program (Long-term Observation and Research of the East China Sea), a Taiwanese contribution to the international SOLAS (Surface Ocean–Lower Atmosphere Study), has begun to study the effects of material transport in the waters of the northwest Pacific Ocean. As part of this program we conducted the first sea-based attempt to study POC export fluxes during or shortly after dust storm events in the subtropical northwest Pacific Ocean.

2. Materials and methods

Sampling was conducted in the subtropical oligotrophic northwest Pacific Ocean, approximately 160 km northeast of Taiwan (123.15°E, 25.10°N, water depth=1690 m, Fig. 1) from the R/V Ocean Researcher II (OR II), which conducted four discrete cruises in 2007: 15–16 March, 19–23 March, 27 March–1 April, and 9–13 April. Wind speed (hourly average) was measured at Pengjiayu Island (101 m above sea level, Fig. 1) and data were provided by the Central Weather Bureau of Taiwan. Data on aerosol concentrations (suspended particulates <10 µm, abbreviated as PM₁₀) at Matsu Island, which is near China and at a similar latitude to our sampling site (Fig. 1), were provided by the Environmental Protection Administration of Taiwan. Water temperature, density, and salinity were measured using a SeaBird model SBE9/11 with a conductivity–temperature–depth (CTD) recorder. The concentration of nitrate (NO₃) was determined according to Gong et al. (2000). Chl *a* was measured by the relationship between in-situ fluorescence and measured Chl *a*. Sinking particles were collected from 120 m below the euphotic zone by a drifting sediment trap array, which consisted of six 6.8 cm diameter cylindrical plastic core tubes with honeycomb baffles covering the trap mouths (Walsh, 1990; Santschi et al., 2003; Hung et al., 2003, 2004). The array was attached to an electric

surface buoy with a global positioning system (GPS) antenna (TGB-500, TAIYO, Japan). The trap tubes, filled with filtered seawater (Sparkling Clear Polypropylene filter, nominal size 0.5 µm), were deployed daily around 8 am and recovered about 5 pm (so were in the water for about 9 h). Sinking particles were filtered through pre-combusted (500 °C, 6 h) quartz (Whatman QMA) filters. The swimmers caught on the filters were observed using a microscope and carefully removed using forceps. POC concentrations in sinking particles were measured using an elemental analyzer (Elementa, Germany) after filters were HCl-fumed. Suspended particles were sampled using GO-FLO bottles on a rosette at 0, 10, 25, 50, 75, 100, 125, and 150 m depth. POC concentrations in suspended particle samples were measured using an elemental analyzer. The analytical uncertainty (one sigma error) for POC was 2–5% for duplicate measurements. The POC blanks ranged from 1.0 to 1.4 µmol per 25 mm quartz filter, which was wetted with seawater and then rinsed with Milli-Q water. The POC inventory was defined as the depth-integrated water column from the surface to 125 m, which is just deeper than the bottom of the euphotic zone (100–110 m). The POC flux was extrapolated to 24 h (=1 day) on the assumption that the POC flux at night was the same as that during the day.

3. Results and discussion

3.1. Hydrographic settings, dust events, and POC variation during the sampling period

Hydrographic data and NO₃ concentrations within a 50 m depth interval (near the mixed layer depth, MLD, at which the density (σ_θ) increased by 0.1 kg m⁻³ from the σ_θ measured at 10 m; Gong et al., 1999) did not show any change in pattern, but the NO₃ concentration at (or below) 50 m fluctuated after storm events (Fig. 2 and Table 1). For instance, the NO₃ concentration at 50 m was approximately 0.9 µM during the first cruise (15–16 March), decreased to <0.1 µM on 20 March (Fig. 2), was high again

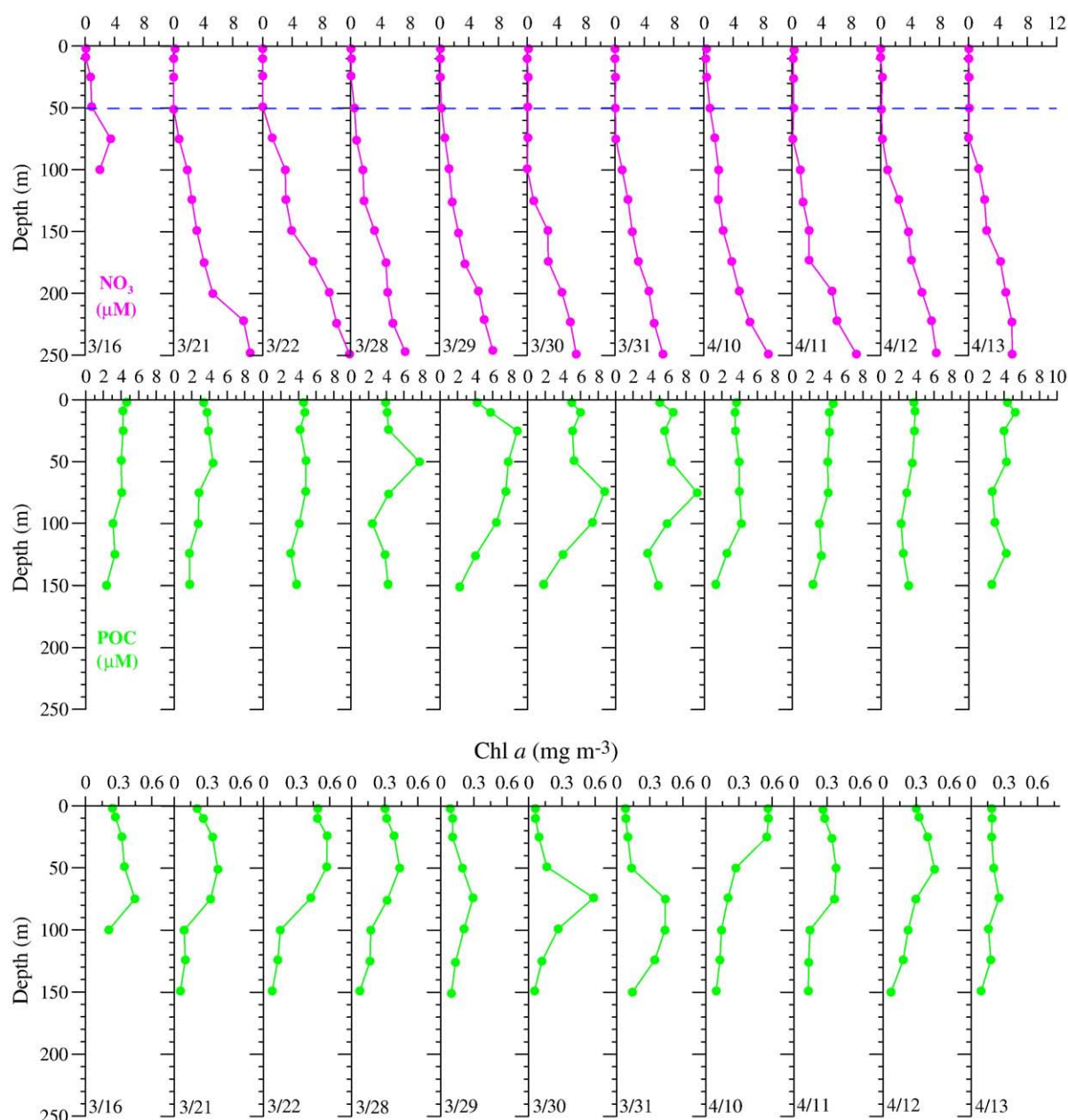


Fig. 2. Vertical distributions of NO_3 concentration (a), POC (b), and Chl *a* (c) in the oligotrophic northwest Pacific Ocean.

($0.5 \mu\text{M}$) on 28 March, then decreased exponentially over 4 days to below the detection limit (Fig. 2 and Table 1). A similar variation in NO_3 concentration in the euphotic zone, especially <50 m, was again observed on 10 April, and a high NO_3 concentration ($0.8 \mu\text{M}$) at 50 m rapidly decreased over 4 days (Table 1). These results demonstrate that subsurface nutrients were entrained in the mixed layer because of vertical water mixing, which may have been driven by strong winds (maximum sustained wind approximately 16.7 m s^{-1} ; Fig. 3 and Table 1). However, nutrients may also have been added to surface waters through enhanced aerosol deposition, but we did not see a marked NO_3 increase in the upper layer (<50 m).

During the first dust storm, which started on 14–15 March (Fig. 3), aerosol concentrations (PM_{10}) at Matsu ranged from 75 to $108 \mu\text{g m}^{-3}$, or about 3–6 times higher than during non-dust storm periods (10 – $20 \mu\text{g m}^{-3}$; Fig. 3). On the night of 16 March a powerful cold front crossed the study area from China. The hourly wind speed increased from 4 m s^{-1} to 12 m s^{-1} , and the ship was forced back to Taiwan. Between 17 and 18 March the study area was subject to severe weather, with wind gusts up to 11 m s^{-1} . During this time elevated POC (Fig. 2) and Chl *a* concentrations (Fig. 2) were observed in the surface layer, and the integrated POC stock (surface to 125 m; 5.9 g m^{-2}) was higher than at previous times during the study, and also in comparison to previously reported levels (3.0 – 3.6 g m^{-2} , Liu et al., 1995; 3.7 g m^{-2} , Hung and Gong, 2007) in spring and autumn under normal weather conditions.

During the second cruise (19–23 March) the winds did not subside until 21 March (gusts up to 9.5 m s^{-1} were recorded on 20 March), but an elevated PM_{10} dust value

($\sim 100 \mu\text{g m}^{-3}$, Fig. 3) was found only between 21 and 22 March. Between 24 and 26 March two major dust storms occurred while we were not at sea, but a third strong dust storm on 29 March (wind speeds 5 – 10 m s^{-1} between 26 March and 1 April; Fig. 3) generated PM_{10} values of approximately $200 \mu\text{g m}^{-3}$ (Fig. 3). Vertical profiles of Chl *a* during the second and third cruises were reflected in elevated NO_3 levels in the surface layer (top 50 m), with peak Chl *a* values to 75 m or deeper suggesting that NO_3 had fueled biomass production at depth (Fig. 2). A similar Chl *a* pattern was observed during the final cruise. The slightly elevated POC concentration in the surface layer, which appeared 1–2 days after dust storm events during the second and third cruises, is likely to have been caused by both nutrient deposition by dust, and entrainment of NO_3 in subsurface waters. On 23 March the POC concentration gradually increased with increasing depth to 50 m, and a similar event was observed during 28–31 March, with POC peak values at both 50 m and 75 m evident after the dust storm events of 26–27 March. These results suggest that enhanced POC concentration was induced by nitrogen input from both atmospheric deposition and wind-induced water column mixing. The largest dust storm during the study period occurred on 2–3 April, but sampling in the resulting conditions was not possible.

During the last cruise, dust storm events on 9–10 April (PM_{10} $\sim 130 \mu\text{g m}^{-3}$) and 11–12 April (PM_{10} $\sim 140 \mu\text{g m}^{-3}$) were accompanied by a medium-strength cold front. The POC distribution pattern was similar to that found during the second and third cruises, with slightly elevated POC concentrations in the surface layer. A comprehensive analysis of the POC inventories (to 125 m depth) during the sampling period revealed

Table 1

Depths of the mixed layer and euphotic zone, and temperature, salinity, NO_3 , integrated Chl a , and POC levels, during four assessment periods in the subtropical northwest Pacific Ocean

Date	MLD (m)	EZ (m)	*T (°C)	*S	* NO_3 (μM)	*In-Chl a (mg m^{-2})	In-POC (g m^{-2})
3/16	64	87	24.247	34.719	0.9	40.86	5.99
3/20	78	86	24.273	34.728	n.a.	n.a.	n.a.
3/21	66	94	24.743	34.661	<0.1	31.18	4.94
3/22	66	80	24.915	34.584	<0.1	50.51	6.58
3/28	60	88	25.058	34.598	0.5	41.94	6.79
3/29	51	95	24.366	34.721	0.3	29.11	10.32
3/30	53	112	25.789	34.730	<0.1	33.33	9.42
3/31	50	105	25.753	34.604	<0.1	28.88	9.60
4/10	72	84	24.462	34.691	0.8	31.68	5.61
4/11	88	81	25.507	34.602	0.3	34.95	5.79
4/12	71	92	25.224	34.667	<0.1	36.75	4.70
4/13	81	96	24.896	34.716	<0.1	27.02	5.60

MLD: mixed layer depth, EZ: euphotic zone, In-Chl a and In-POC: integrated Chl a and POC from 0–125 m, *: values at 50 m (within the MLD), n.a.: data not available.

they were significantly higher (up to 3-fold) than both estimates of the average POC inventory (3.7 g m^{-2}) and previously reported values ($3.0\text{--}3.6 \text{ g m}^{-2}$; Liu et al., 1995), indicating that POC inventories taken in spring are much higher than average POC values (Fig. 3).

3.2. POC export flux

The export flux of POC ranged from 49 ± 1 to $98 \pm 9 \text{ mg m}^{-2} \text{ day}^{-1}$, with the highest POC fluxes ($>49 \text{ mg m}^{-2} \text{ day}^{-1}$) being found during the second, third, and final cruises (Fig. 3). Dust deposition during the dust storm on 14–15 March was followed 2 days later by an enhanced POC flux ($79 \pm 7 \text{ mg m}^{-2} \text{ day}^{-1}$), almost three times greater than that reported previously ($28 \pm 1 \text{ mg m}^{-2} \text{ day}^{-1}$) for the subtropical northwest Pacific Ocean (Hung and Gong, 2007). These findings suggest that enhanced POC flux may be induced by dust storms, or by enhanced vertical mixing bringing a supply of nutrients

and other metals into the euphotic zone. Dissolved Fe and nutrients, such as NO_3 and ammonium, are always found in aerosols during dust storm events (Gong et al., 2005). Thus, the high POC flux we measured during the first cruise may have been caused by the coupling of dust deposition and wind-induced mixing of the water column.

The high export flux of POC ($87 \pm 1 \text{ mg m}^{-2} \text{ day}^{-1}$) found on the second cruise occurred on 20 March, 3–4 days after very stormy weather. There was also an elevated peak in atmospheric dust from 21–22 March near the study area. This suggests that the high POC export flux arose because of enhanced vertical mixing of subsurface water, caused by the strong winds. This enhanced mixing could have brought nutrient-rich water to the euphotic zone. The concentrations of NO_3 ($<0.1 \mu\text{M}$ at 50 m) may not offer support for this mechanism (Table 1), but the cold water ($<25^\circ\text{C}$) found at 50 m suggests that nutrients were brought to the surface from the colder and nutrient-enriched waters below (Gong et al., 1997). However, this explanation is hypothetical, in that we cannot explain the high POC export flux based only on our collected data. After the elevated export flux event occurred, the POC flux gradually decreased to $49 \pm 1 \text{ mg m}^{-2} \text{ day}^{-1}$ within 2 days (by 22 March). Over the same time the POC inventory increased from 4.9 to 6.6 g m^{-2} , suggesting that net phytoplankton growth increased POC standing stocks, which may have subsequently declined because of particle sinking or zooplankton grazing.

On the third cruise (27 March–1 April), the POC flux gradually increased from $60 \pm 6 \text{ mg m}^{-2} \text{ day}^{-1}$ on 28 March to a maximum of $85 \pm 2 \text{ mg m}^{-2} \text{ day}^{-1}$ on 30 March. A severe dust storm occurred on 25–27 March, and 2–3 days later there was an increase in both the POC flux and the POC inventory in these oligotrophic waters. In contrast to our area, in high nutrient–low chlorophyll areas the POC export flux responses to dust storms take longer (about a week) (Bishop et al., 2002; Honda, 2003). In such areas iron from dust storms may greatly influence the growth of phytoplankton. However, in oligotrophic waters, such as in our study area, nutrients are depleted, and phytoplankton mainly consist of nanoplankton and picoplankton. In our study, if wind resulted in nutrients becoming available in ambient seawater, either through dust deposition or atmospherically-induced mixing of nutrients into subsurface waters (Fig. 2 and Table 1), phytoplankton may have rapidly utilized the micronutrients, resulting in an increase in POC export flux. Remarkably, the two areas (the high nutrient–low chlorophyll zones and the oligotrophic oceans) respond differently to nutrient inputs. One explanation for this is that the major phytoplankton assemblages in the subtropical northwest Pacific Ocean (i.e., oligotrophic waters) are small picoplankton including *Synechococcus* and *Prochlorococcus* (Chang et al., 1996; Chiang et al., 2002; Chang et al., unpublished data), but the major phytoplankton component in the high nutrient–low chlorophyll areas are larger, including diatoms (Boyd et al., 2004, 2005). Picoplankton can grow faster than microplankton because of their higher photosynthesis rates. Moreover, because NO_3 is present in

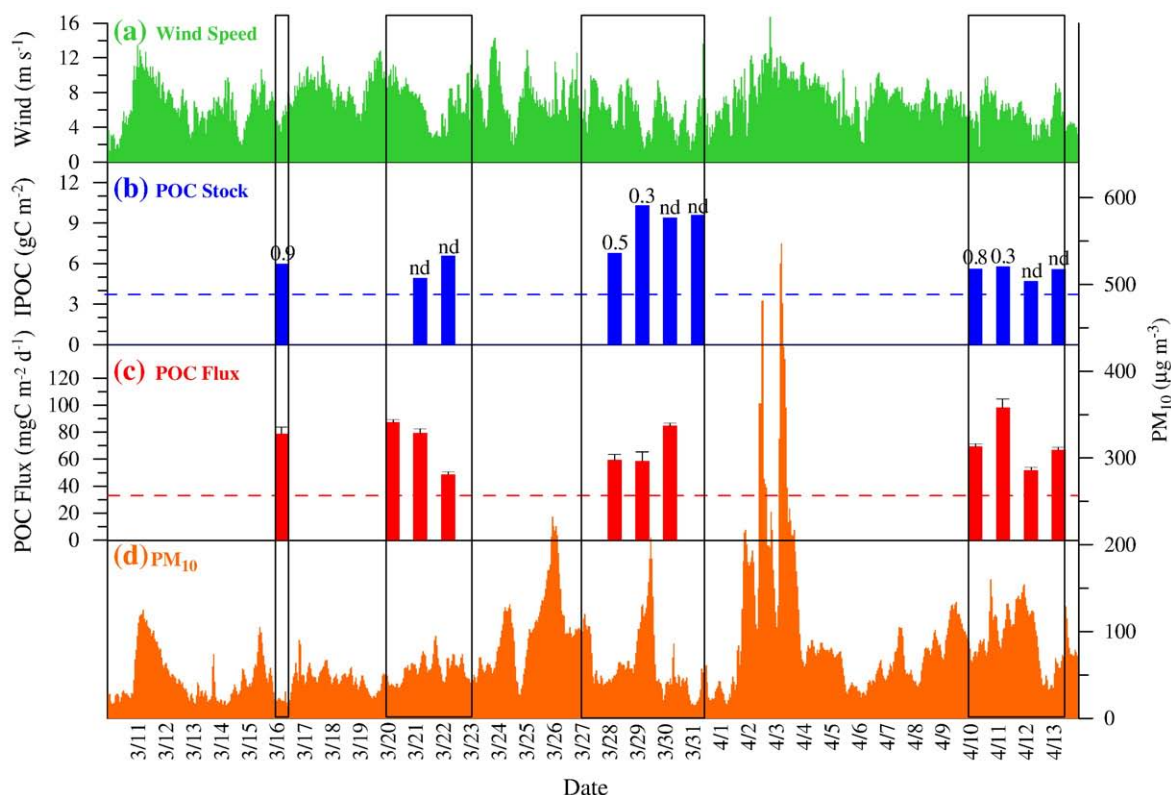


Fig. 3. (a) Wind speed measurements at Pengjiayu Island. Each data point represents a 1-hour average value; (b) variation in POC inventories at the sampling location. The blue dashed line represents data from Hung and Gong (2007); (c) export fluxes of POC at the sampling station. The red line represents the estimated POC export flux in the oligotrophic northwest Pacific Ocean (Hung and Gong, 2007); and, (d) concentrations of aerosols (PM_{10}) at Matsu (close to China). The vertical lines separating the data represent the first (16–17 March), second (19–23 March), third (26–31 March), and fourth (10–13 April) cruises, respectively (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.).

approximately micromolar concentrations and iron concentrations are nanomolar or less, phytoplankton can take up the NO_3 more quickly than the iron, based on the Michaelis–Menten equation (Voet and Voet, 1995). Furthermore, phytoplankton usually take up the more bioavailable dissolved iron, rather than particulate iron. According to a recent report of Hsu et al. (in press), particulate Fe has a solubility of <20%. However, the nitrogen sources (i.e., NO_3 and ammonium) are almost exclusively in dissolved forms. As a consequence, NO_3 can be taken up more rapidly than dissolved iron.

During the final cruise (9–13 April), a POC flux of $69 \pm 2 \text{ mg m}^{-2} \text{ day}^{-1}$ was detected on 10 April; this peaked at $98 \pm 9 \text{ mg m}^{-2} \text{ day}^{-1}$ on 11 April and dropped to $66 \pm 1 \text{ mg m}^{-2} \text{ day}^{-1}$ on 13 April. Remarkably, both the POC inventory and the POC flux during the final cruise were lower than on the third cruise. This could have been because the storm was weak and the dust moderate. Increased grazing may also result in a decline in POC inventory, and cause considerable POC flux. Overall, the POC flux in the subtropical northwest Pacific Ocean during dust storm periods ranged from 49 to $98 \text{ mg C m}^{-2} \text{ day}^{-1}$ (average $71 \text{ mg C m}^{-2} \text{ day}^{-1}$), which is about 2.5-fold higher than at three comparable subtropical ocean sites (20–30 $\text{mg C m}^{-2} \text{ day}^{-1}$; ALOHA, BATS, and ESTOC; Neuer et al., 2002).

3.3. Fe and nitrogen input during the dust storm period

Variations in POC flux in the subtropical northwest Pacific Ocean can be attributed to a combination of the northeast monsoon and dust storms, but the influences of these two weather events are difficult to distinguish. We used a simple calculation to estimate the contribution of aerosol iron fertilization and nitrogen inputs to phytoplankton growth. This calculation relies on the assumption that the average dust concentration and the average dry deposition velocity for iron are $100 \mu\text{g m}^{-3}$ and 1 cm s^{-1} , respectively (Natalie et al., 2005). If the average Fe content in aerosol particles is 3% (w/w), and the soluble Fe fraction is 10%, the soluble Fe input will be $2.6 \times 10^2 \mu\text{g Fe m}^{-2} \text{ day}^{-1}$ (Natalie et al., 2005). The C:Fe ratio in marine phytoplankton can range from 10^2 :5 to 10^3 :10, if Redfield ratios and reported metal contents of marine phytoplankton are accurate (Redfield et al., 1963; Morel and Hudson, 1985; Bruland et al., 1991). Using an average C:Fe molar ratio of 10,000:1 in the calculation, the estimated phytoplankton growth (PP) would be $600 \text{ mg C m}^{-2} \text{ day}^{-1}$ under nutrient-replete condition. This value is within the primary production (PP) range (260 – $1000 \text{ mg C m}^{-2} \text{ day}^{-1}$) recently determined by Gong et al. (2005) in a study in this area, suggesting that estimates of phytoplankton growth based on a C:Fe approach may be feasible. However, because the subtropical oligotrophic northwest Pacific Ocean is a nitrogen-limited, and not an Fe-limited, system, this calculation may overestimate the importance of dust storm inputs. It is difficult to estimate, based on variable wind speed and direction, the quantity of nutrients made available to plankton through vertical mixing. An alternative approach is to compute the combined contribution of nitrogen via dry dust storm deposition and the wet deposition flux. Hsu et al. (in press) reported the chemical composition of particulate matter on dust collected during the OR-II cruises noted in this study. The dry inorganic nitrogen (i.e., the sum of NO_3 and ammonium) flux was about $0.056 \pm 0.067 \text{ mmol N m}^{-2} \text{ day}^{-1}$, which is consistent with the results of Nakamura et al. (2005) but slightly less than reported by Chen and Chen (2008). With respect to the wet deposition flux of nitrogen, we estimated wet nitrogen deposition based on simulated data of monthly mean NO_3 concentrations in precipitation over East Asia (e.g., An et al., 2002), and monthly rainfall data (207 mm in March and 149 mm in April, 2007) collected from the meteorological station on Pengjia Island (<http://www.cwb.gov.tw/>). The total of rainy days in March and April 2007 were approximately 15 and 13 days, respectively. Thus, the average daily rainfall on Pengjia Island was 11.5 – 13.8 mm day^{-1} (average 13 mm day^{-1}). The monthly mean NO_3 concentration in precipitation over East Asia is about 3 mmol m^{-3} . The estimated wet deposition of nitrogen is thus $0.04 \text{ mmol N m}^{-2} \text{ day}^{-1}$, and the combined nitrogen (dry and wet) deposition is thus approximately $0.1 \text{ mmol N m}^{-2} \text{ day}^{-1}$ (i.e., $1.4 \text{ mg N m}^{-2} \text{ day}^{-1}$).

The estimated nitrogen contributed from both dry and wet deposition within the euphotic zone (0–120 m) is $0.07 \mu\text{M}$ when a residence time (based on the calculation of nitrogen inventory and nitrogen flux = POC flux/6.6) of 88 days is adopted. This is significantly lower than the observed NO_3 concentration within the mixed layer (range: undetectable to 0.5 – $0.9 \mu\text{M}$ at 50 m; average: $0.7 \mu\text{M}$) of the euphotic zone. This suggests that

the nitrogen contribution from both dry and wet deposition is significantly lower than that from sub-surface water entrainment. If an analogous approach is applied to Fe deposition, the input of N from both dry and wet deposition would be $1.4 \text{ mg N m}^{-2} \text{ day}^{-1}$. As calculated earlier, the average export flux of N through the base of the euphotic zone is $14.1 \text{ mg N m}^{-2} \text{ day}^{-1}$ (assuming C:N=6.6), which accounts for only 10% of the N export flux relative to the maximum nitrogen input from both dry and wet deposition. The integrated NO_3 concentration within the mixed layer depth (MLD), the actual integrated NO_3 -produced POC, and the estimated POC flux during the sampling periods 16–22 March, 28–31 March, and 10–13 April are shown in Table 2. For example, the actual (integrated from 0 to 64 m) and estimated NO_3 standing stocks ($0.9 \mu\text{M} \times 64 \text{ m}$) within the MLD (64 m) on 16 March were 0.039 and $0.058 \text{ mol N m}^{-2}$, respectively. The estimated POC production was about 1451 mg C m^{-2} , and the average conservative particle loss (i.e., estimated POC flux) was $207 \text{ mg C m}^{-2} \text{ day}^{-1}$ (Table 2). A similar situation (i.e., estimated nitrate standing stock > actual nitrate standing stock) occurred on 28–31 March and 10–13 April. An elevated POC flux was not observed immediately after the dust storm event, but did occur approximately 1–2 days after dust events accompanied by strong winds (Fig. 3). Conversely, we did not find a positive correlation between change in NO_3 stock (the difference between the integrated NO_3 within the MLD, and the actual integrated NO_3) and POC fluxes in the study area, which implies that (A) NO_3 consumption is not directly proportional to the POC flux during periods when NO_3 is entrained into the euphotic zone, and, (B) an enhanced POC flux evident 1–2 days after a dust storm event reflects a short time lag. Overall, the average measured POC flux ($78 \text{ mg C m}^{-2} \text{ day}^{-1}$) from the sampling period 16–22 March was significantly less than the estimated POC flux ($207 \text{ mg C m}^{-2} \text{ day}^{-1}$; Table 2). These findings demonstrate that most of the newly-produced NO_3 -supported phytoplankton POC was not directly exported within the sampling period, and the deficit could be because of bacterial re-mineralization and/or phytoplankton growth. Assessment of the latter is a key challenge in understanding the fate of carbon fixed by phytoplankton in different oceanic environments. To date, the fate of carbon taken up by phytoplankton (including iron-induced phytoplankton blooms) and the efficiency with which the carbon is exported into the ocean interior, remain unclear (see Boyd et al., 2004 and references therein).

According to the above estimates, vertical water mixing must be the primary mechanism delivering nutrients into the euphotic zone in the subtropical oligotrophic northwest Pacific Ocean. It is difficult to evaluate actual phytoplankton growth based on individual dust storm events, because dust and associated nutrient concentrations are variable, and particle sizes in aerosols are irregular. However, estimates of the order of magnitude calculated by us above are suggestive of the importance of storm events in enhancing vertical mixing and POC flux, and hence removal of atmospheric CO_2 from the ocean surface.

4. Conclusions

In summary, this study is the first to document that atmospheric forcing, brought about by the strong northeast monsoon and dust storm events, induced a POC export flux that was 2–3-fold higher than that seen during non-dust storm periods, in the subtropical oligotrophic waters of the northwest Pacific Ocean. The POC export flux was enhanced mainly by increased nutrients that were made available through wind-induced vertical mixing of subsurface water; atmospheric nutrient inputs during dust storms could not alone explain the enhancement. Although we are currently unable to precisely estimate, from storm wind strength, the contribution of dust input to phytoplankton growth, or to quantify the relationship between POC export flux enhancement and wind speed, the results of this study provide qualitative evidence of the importance of such events. Further research is needed to characterize and quantify the importance of storm events to long-term POC export flux in oligotrophic waters.

Acknowledgments

We are grateful to J.Z. Tseng, W.Z. Yang, P.J. Hsu, and the crew of the R/V Ocean Research II for their assistance in collecting samples. We also thank Dr. E.D. Peltzer, the Associate Editor, and two anonymous reviewers who gave constructive comments that improved the paper. This research was funded by grants from the National Science Council (NSC 95-2611-M-019-020-MY3, NSC 95-2611-M-019-021-MY3, NSC96-2611-M-019-011, NSC97-2745-M-019-001) and the Center for Marine Bioscience and Biotechnology at National Taiwan Ocean University.

References

- An, J., Udeh, H., Wang, Z., Matsuda, K., Kajino, M., Cheng, X., 2002. Simulations of monthly mean nitrate concentrations in precipitation over East Asia. *Atmos. Environ.* 36, 4159–4171.

Table 2

Standing stock of NO_3 and estimated POC flux during sampling periods (16–22 March, 28–31 March, and 10–13 April 2007)

Date	Est. NO_3 (mol N m^{-2})	Actual NO_3 (mol N m^{-2})	Lost NO_3 (mol N m^{-2})	Est. POC (mg C m^{-2})	E. POC flux ($\text{mg C m}^{-2} \text{ day}^{-1}$)	M. POC flux ($\text{mg C m}^{-2} \text{ day}^{-1}$)
3/16	0.058	0.039	0.018	1451	207	78
3/28	0.032	0.019	0.012	983	246	60
4/10	0.057	0.047	0.010	777	194	70

Est. NO_3 : the estimated NO_3 stock within the mixed layer.

Actual NO_3 : the actual integrated NO_3 stock within the mixed layer.

Lost NO_3 : the difference between the estimated and actual integrated NO_3 stock.

Est. POC: the estimated POC stock using the Redfield ratio (C = $6.6 \times$ nitrate stock).

Est. POC flux: the estimated POC flux (on 16 March: Est. POC stock/7 days (16–22 March); on 28 March: Est. POC stock/4 days (28–31 March); on 10 April: Est. POC stock/4 days (10–13 April)).

M.POC flux: the POC flux measured using sediment traps.

- Bishop, J.K., Davis, R.E., Sherman, J.T., 2002. Robotic observations of dust storm enhancement of carbon storm enhancement of carbon biomass in the North Pacific. *Science* 298, 817–821.
- Boyd, P.W., Law, C.S., Wong, C.S., Hori, Y., Tsuda, A., Levasseur, M., Takeda, S., Rivkin, R., Harrison, P.J., Strzepek, R., Gower, J., McKay, R.M., Abraham, E., Arychuk, M., Barwell-Clarke, J., Crawford, W., Crawford, D., Hale, M., Harad, K., Johnson, K., Kiyosawa, H., Kudo, I., Marchetti, A., Miller, W., Needoba, J., Nishioka, J., Ogawa, H., Page, J., Robert, M., Sato, H., Sastri, A., Sherry, N., Soutar, T., Sutherland, N., Taira, Y., Whitney, F., Wong, S.-K. E., Yoshimura, T., 2004. The decline and fate of an iron-induced subarctic phytoplankton bloom. *Nature* 428, 549–553.
- Boyd, P.W., Strzepek, R., Takeda, S., Jackson, G., Wong, C.S., McKay, R.M., Law, C., Kiyosawa, H., Saito, H., Sherry, N., Johnson, K., Gower, J., Ramaiah, N., 2005. The evolution and termination of an iron-induced mesoscale bloom in the northeast subarctic Pacific. *Limnol. Oceanogr.* 50, 1872–1886.
- Bruland, K.W., Donat, J.R., Hutchins, D.A., 1991. Interactive influences of bioactive trace metals on biological production in oceanic waters. *Limnol. Oceanogr.* 36, 1555–1577.
- Chang, J., Chung, C.-C., Gong, G.-C., 1996. Influences of cyclones on chlorophyll a concentration and *Synechococcus* abundance in a subtropical western Pacific coastal ecosystem. *Mar. Ecol. Prog. Ser.* 140, 199–205.
- Chen, H.-Y., Chen, L.-D., 2008. The importance of anthropogenic inputs and continental-derived dust for the distribution and flux of water-soluble nitrogen and phosphorus species in aerosol within the atmosphere over the East China Sea. *J. Geophys. Res.* 113, D11303. doi:10.1029/2007JD009491.
- Chiang, K.-P., Kuo, M.-C., Chang, J., Wang, R.-H., Gong, G.-C., 2002. Spatial and temporal variation of the *Synechococcus* population in the East China Sea and its contribution to phytoplankton biomass. *Cont. Shelf Res.* 22, 3–13.
- DiTullio, G.R., Laws, E.A., 1991. Impact of an atmospheric-oceanic disturbance on phytoplankton community dynamics in the North Pacific Central Gyre. *Deep-Sea Res.* A 38, 1205–1329.
- Gong, G.-C., Shiah, F.-K., Liu, K.-K., Chuang, W.-S., Chang, J., 1997. Effect of the Kuroshio intrusion on the chlorophyll distribution in the southern East China Sea north of Taiwan during spring, 1993. *Cont. Shelf Res.* 17, 79–94.
- Gong, G.-C., Chang, J., Wen, Y.-H., 1999. Estimation of annual primary production in the Kuroshio waters northeast of Taiwan using a photosynthesis-irradiance model. *Deep-Sea Res.* 46, 93–108.
- Gong, G.-C., Shiah, F.-K., Liu, K.-K., Wen, Y.-H., Liang, M.-H., 2000. Spatial and temporal variation of chlorophyll a, primary productivity and chemical hydrography in the southern East China Sea. *Cont. Shelf Res.* 20, 411–436.
- Gong, G.-C., Hsu, S.-J., Chen, H.-Y., 2005. The effects of Asian dust on biological productivity in the oligotrophic Kuroshio waters of the subtropical northwest Pacific Ocean. Workshop on the marine environment in the East Asian Marginal Seas-transport of materials. Kyushu University, Kasuga, Fukuoka, Japan, pp. 34–38.
- Herut, B., Zohary, T., Krom, M.D., Mantoura, R.F.C., Pitta, P., Psarra, S., Rassoulzadegan, F., Tanaka, T., Thingstad, T.F., 2005. Response of East Mediterranean surface water to Saharan dust: on-board microcosm experiment and field observations. *Deep-Sea Res.* 52, 3024–3040.
- Honda, M., 2003. Biological Pump in Northwestern North Pacific. *J. Oceanogr.* 59, 671–684.
- Hsu, S.-C., Wong, G.T.F., Gong, G.-C., Shiah, F.-K., Huang, Y.-T., Kao, S.-J., Tsai, F., Lung, S.-C.C., Lin, F.-J., Lin, I.-I., Hung, C.-C., Tseng, C.-M. in press. Sources, solubility, and dry deposition of aerosol trace elements over the East China Sea. *Mar. Chem.*
- Hung, C.-C., Gong, G.-C., 2007. Export flux of POC in the main stream of the Kuroshio. *Geophys. Res. Lett.* 34, L14610. doi:10.1029/2007GL029633.
- Hung, C.-C., Guo, L., Schultz, G., Pinckney, J., Santschi, P.H., 2003. Production and flux of carbohydrate species in the Gulf of Mexico. *Glob. Biogeochem. Cycles* 17. doi:10.1029/2002GB001988.
- Hung, C.-C., Guo, L., Roberts, K., Santschi, P.H., 2004. Upper ocean carbon flux determined by ^{234}Th and sediment traps in the Gulf of Mexico. *Geochim. J.* 38, 601–611.
- Lin, I.-I., Liu, W.-T., Wu, C.-C., Wong, G.T.F., Hu, C., Chen, Z., Liang, W.-D., Yang, Y., Liu, K.K., 2003. New evidence for enhanced ocean primary production triggered by tropical cyclone. *Geophys. Res. Lett.* 30 (3), 1131. doi:10.1029/2002GL015674.
- Liu, K.K., Lai, Z.-L., Gong, G.-C., Shiah, F.-K., 1995. Distribution of particulate organic matter in the southern East China Sea: implications in production and transport. *Terrest. Atmos. Ocean. Sci.* 6, 27–45.
- Morel, F.M.M., Hudson, R.J.M., 1985. The geobiological cycle of trace elements in aquatic systems: Redfield revisited. In: Stumm, W. (Ed.), *Chemical Processes in Lakes*. John Wiley, pp. 251–281.
- Nakamura, T., Matsumoto, K., Uematsu, M., 2005. Chemical characteristics of aerosols transported from Asia to the East China Sea: an evaluation of anthropogenic combined nitrogen deposition in autumn. *Atmos. Environ.* 39, 1749–1758.
- Natalie, M.M., Baker, A.R., Bergametti, G., Brooks, N., Duce, R.A., Jickells, T.D., Kubilary, N., Prospero, J.M., Tegen, I., 2005. Atmospheric global dust cycle and iron inputs to the ocean. *Glob. Biogeochem. Cycles* 19, GB4025. doi:10.1029/2004GB002402.
- Neuer, S., Davenport, R., Freudenthal, T., Wefer, G., Llinas, O., Rueda, M.-J., Steinberg, D.K., Karl, D.M., 2002. Differences in the biological carbon pump at three subtropical ocean sites. *Geophys. Res. Lett.* 29, 1885. doi:10.1029/2002GL015393.
- Redfield, A.C., Ketchum, B.H., Richards, R.A., 1963. The influence of organisms on the composition of sea-water. In: Hill, M.N. (Ed.), *The Sea*. Interscience, pp. 26–77.
- Santschi, P.H., Hung, C.-C., Guo, L., Schultz, G.G., Pinckney, J., Walsh, I., 2003. Control of acid polysaccharide production, ^{234}Th and particulate organic carbon export flux by marine organisms. *Geophys. Res. Lett.* 30, 1044. doi:10.1029/2002GL016046 (2003).
- Sun, J., Zhang, M., Liu, T., 2001. Spatial and temporal characteristics of dust storms in China and its surrounding regions, 1960–1999: relations to source area and climate. *J. Geophys. Res.* 106, 10325–10333.
- Uematsu, M., Toratani, M., Kajino, M., Narita, Y., Senga, Y., Kimoto, T., 2004. Enhancement of primary productivity in the western North Pacific caused by the eruption of Miyakejima Volcano. *Geophys. Res. Lett.* 31, L06106. doi:10.1029/2003GL018790.
- Walsh, I., 1990. Project CATSTIX: Camera, transmissometer, and sediment trap integration experiment, Ph.D. dissertation, Texas A&M University, College Station, TX, 1990.
- Wells, M.L., 2003. The level of iron enrichment required to initiate diatom blooms in HNLC waters. *Mar. Chem.* 82, 101–114.
- Wen, L.S., Jiann, K.T., Santschi, P.H., 2006. Physico-chemical speciation of bioactive trace metals (Cd, Cu, Fe, Ni) in the oligotrophic South China Sea. *Mar. Chem.* 101, 104–129.
- Wu, J.F., Chung, S.W., Wen, L.S., Liu, K.K., Chen, Y.L.L., Chen, H.Y., Karl, D.M., 2003. Dissolved inorganic phosphorus, dissolved iron, and *Trichodesmium* in the oligotrophic South China Sea. *Global Biogeochem. Cycles* 17. doi:10.1029/2002GB001924.
- Yuan, W., Zhang, J., 2006. High correlations between Asian dust events and biological productivity in the western North Pacific. *J. Geophys. Res.* 33, L07603. doi:10.1029/2005GL025174.