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南海生地化整合研究－子計畫五
南海新生產力－漂 浮式沈降顆粒收集器之應用

New Production in the South China Sea: Floating Trap Application

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台灣西南海域顆粒有機碳輸出通量: 釷－234應用

摘　要

本研究在台灣西南海域兩測站採集大體積海水樣，進行溶解態和顆粒態釷－234分析。經由不同採樣梯次，本研究亦量測了顆粒態有機碳濃度和其他水文化學參數的垂直剖面圖。由測站水體中大量不足釷－234顯示本研究海域受到快速的清除及顆粒移除作用影響。由釷－234/鈾－238不平衡計算結果顯示兩測站釷－234垂直通量由近表水的150 dpm/m^2/d隨深度遞增；在水深300公尺處，測站B2和測站HT的釷－234垂直通量分別達5800和8200 dpm/m^2/d。受到較大的陸源輸入影響，測站HT釷－234垂直通量隨深度的增加率較測站B2高50 %。

利用釷－234/鈾－238不平衡和懸浮顆粒中有機碳及釷－234比值關係，本研究估算出台灣西南海域的顆粒有機碳輸出通量。於兩測站的透光層底部，顆粒有機碳輸出通量達20~50 mM/m^2/d；在水深300公尺處，測站B2和測站HT的釷－234垂直通量則達約75 mM/m^2/d。在次表層水體中，測站HT的顆粒有機碳輸出通量較測站B2高，且有一明顯的極大值。測站HT的顆粒有機碳輸出通量極大值伴隨著亞硝酸極大值。與釷－234垂直通量不同，測站HT兩組採樣梯次所得之顆粒有機碳輸出通量顯現極大的差異性；與晚上八時比較，下午兩點顆粒有機碳輸出通量顯現較高的極大值和較高的隨深度遞減率，此現象可能因上層水體中顆粒有機碳產量和呼吸率的日變化所造成的結果。

Export Flux of Particulate Organic Carbon from the Water Column off Southwestern Taiwan: ^{234}Th Elucidation

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Abstract

Large-volume seawater samples were collected from the water column at two stations off southwestern Taiwan for the measurements of dissolved and particulate ^{234}Th. Parallel vertical profiles of particulate organic carbon and other hydrographic parameters were also determined from separate sampling casts. Enhanced scavenging and removal processes are evident in the study area as indicated by large deficiencies of ^{234}Th relative to ^{234}U throughout the whole water column. Vertical ^{234}Th fluxes calculated from the ^{234}Th-^{238}U disequilibria increase monotonically from ~150 dpm/m^2/d in the surface at both stations to 5800 and 8200 dpm/m^2/d at the depth of 300 m of station B2 and station HT, respectively. The increasing rate of ^{234}Th fluxes with depth at station HT is 50% higher than that of station B2 due to shoreward intensification of scavenging force originated from terrestrial input.

Export fluxes of particulate organic carbon in the coastal regime off southwest Taiwan were estimated from ^{234}Th-^{238}U disequilibria and POC/^{234}Th ratios in suspended particles in the water column. The calculated export fluxes of POC at the two stations range from 20 to 50 mM/m^2/d at the base of euphotic layer and converge to ~75 mM/m^2/d at 300 m. In the subsurface layer, station HT shows a higher POC flux than station B2. At station HT, a distinctive maximum of POC fluxes were found in the layer enriched with nitrite. Unlike ^{234}Th fluxes, POC fluxes obtained by two sampling casts, separated by ~18 hours, at station HT reveal different pattern. Samples taken from the afternoon (2 p.m.) show a higher value at the maximum and sharper decreasing rate with depth than those taken in the evening (8 p.m.), which may be resulted from diurnal variation of POC production and respiration rates in the upper water column.

Introduction

In order to understand the cycling process of carbon in the ocean, it is crucial to know the amount of organic carbon settled out the euphotic layer, i.e., export production. Export production provides an important constraint of carbon balance in the euphotic layer and balanced by the production supported by upwelled nutrients and other external nutrient sources. Vertical settling of particulate organic carbon serves as an important

pathway of this exported production and can either be directly measured by sediment trap deployed at the base of euphotic layer or estimated from ^{234}Th-^{238}U disequilibria, a powerful tool to trace carbon in the upper ocean (IOC, 1994; Buesseler, 1998). Recently, these two approaches have widely been used in various oceanic regimes, including open ocean (Buesseler et al., 1995; Bacon et al., 1996; Murray et al., 1996) and coastal ocean (Baskaran et al., 1996; Santschi et al., 1999; Hall et al., 2000).

Th-234 ($t_{1/2}$=24.1 days) is continuously produced from ^{238}U decay and is one of the most particle-reactive elements in seawater. The ^{234}Th-^{238}U disequilibria can quantitatively estimate the scavenging and removal rates by simple mass balance calculation if both dissolved and particulate ^{234}Th were measured (Coale and Bruland, 1985; Wei and Murray, 1992), and the removal rates of associated elements in the particle can then be estimated by multiplying ^{234}Th flux to the ratio of ^{234}Th and elements interested.

Outskirting northeastern South China Sea, the study area is influenced by the intrusion of Kuroshio Current through Bashi Channel (Shaw, 1989). The region has been used as a test site for various experiments. Floating trap experiments were successfully carried out in this region and the ^{234}Th fluxes measured by the floating trap were consistent with that estimated from the ^{234}Th/^{234}U disequilibria in seawater, ranging from 363 to 2290 dpm/m^2/d (Wei et al., 1994). In this paper, we will present results of ^{234}Th and particulate organic carbon (POC) measurements from two stations off southwestern Taiwan and give an estimate of export flux of particulate organic carbon from the water column.

Methods

Seawater samples were collected at two stations shown in Figure 1 during 28~31 March 1999, onboard R/V Ocean Researcher I (cruise # 545). A CTD/20 l Go-Flo system attached with a fluorescence sensor was used to collect large volume seawater for ^{234}Th determination. At station HT (Figure 1), a total of 12 CTD/Rosette casts were repeated in the time span of 33 hours, among which two casts (cast #8 at 8 p.m. of 29 March and #26 at 2 p.m. of 30 March) were used for dissolved and particulate ^{234}Th measurements. Seawater was immediately pressure-filtered by compressed air through a

pre-weighed 142 mm Nuclepore filter (0.45 μm) mounted in a Plexiglas filter holder.

Filtered seawater (~20 l) was transferred into a cubitainer, acidified with approximately 20 ml of concentrated HCl and spiked with 35 dpm ^{230}Th yield tracer as well as 60 mg Fe carrier. The samples were then aerated for at least 4 hours to help achieve isotopic equilibrium (Wei and Hung, 1993). Without interrupting the aeration, 12 N NaOH was added to raise the pH to 8. The $Fe(OH)_3$ precipitates, with adsorbed thorium, were collected by siphoning and centrifuging, and then dissolved in concentrated HCl to make the samples 9 N HCl. These samples were then passed through an anion exchange column (AG1X-8) preconditioned by 9N HCl to separate uranium from thorium. Thorium samples were purified by passing the sample through three anion exchange columns pre-conditioned with 8 N HNO_3. The sample was evaporated down to one drop and was ready for extraction. Th-234 and the yield tracer, ^{230}Th, were extracted into a 0.4 M TTA (thenoyltrifluoroacetone)-benzene solution and stippled on a stainless-steel disc. Preconcentration and separation of uranium and thorium from the filtered seawater samples were completed in three days after samples were collected.

After weighing for the determination of TSM concentration, the filters were decomposed in the laboratory by soaking in ~10 ml of concentrated NH_4OH. The samples were gently heated to evaporate the NH_4OH then fluxed in $HClO_4$/HF to thoroughly digest organic and inorganic materials. After digestion, the samples were purified and mounted on stainless-steel discs following the same procedures as for dissolved ^{234}Th samples.

The activities of ^{234}Th were counted by a low background (< 0.3 cpm) anticoincidence counter (Tennelec LB-5100) via its β-emiting daughter ^{234}Pa. Blank determinations of all reagents and filter paper used for this study indicated negligible contribution. Chemical yield of thorium was estimated by counting spiked ^{230}Th using silicon surface-barrier detectors (EG&G Ortec 576). The counting efficiencies of the a detectors were calibrated against NIST traceable ^{241}Am (Isotope Products Laboratory 387-67-2-2) and ^{230}Th (Isotope Products Laboratory 387-67-3) standard plates. Activities of ^{234}Th reported here were corrected back to the sampling time after the ingrowth of ^{234}Th from ^{238}U was subtracted.

A separate cast of CTD/Rosette assembly was deployed to collect seawater for POC measurement. Particulate organic carbon was measured on the particulate matter retained by a Whatman 25 mm GF/F glass fiber filter, which was pre-heated at 450°C to reduce background carbon. A Heraeus CHN Rapid Elemental Analyzer was used to measure total organic content after carbonate carbon was removed by adding 1 N HCl solution. The precision of the POC data is ±0.2 μM (Liu et al., 1995).

Results and Discussions

Depth, salinity, calculated ^{238}U activities from Ku et al. (1977), concentrations of total suspended matter (TSM), dissolved, particulate ^{234}Th activities, and POC concentrations are given in Table 1. Uncertainties of all radioisotope data listed were estimated according to the propagation of counting error (± 1σ). Hydrographic and nutrients data are not listed but are available upon request.

The vertical distributions TSM concentration, dissolved and particulate ^{234}Th at stations HT and B2 are shown in Figures 2 and 3, respectively. Total ^{234}Th activities as the sum of dissolved and particulate ^{234}Th are also included in the figures. Dotted lines drawn in the figures represent calculated ^{238}U activities. Similar to the previous results obtained in the vicinity (Hung and Wei, 1992; Wei et al., 1994) and in other coastal regimes (Baskaran, 1996; Santschi et al. 1999), all profiles show the deficiency of ^{234}Th relative to ^{238}U throughout the whole water column, indicating dynamic scavenging phenomenon in the region. The degree of ^{234}Th deviation from secular equilibrium is more pronounced at station HT than station B2, due to enhanced particle scavenging landward. Average residence times of ^{234}Th in the euphotic zone of station HT are 38 and 20 days, with respect to scavenging and particle removal rates, respectively. Comparing to station HT, a longer residence time of dissolved ^{234}Th, 64 days, relative to scavenging rate was found in the euphotic zone of station B2 but similar particulate residence time of 21 days was found. It is noted that there is little difference between two sets of profiles obtained from two casts separated by 18 hours at station HT, as a result of a fast turn over rate of ^{234}Th relative to physical mixing in the study area. For all samples taken, ^{234}Th is dominated by dissolved phase, which represents 60~90 % of total ^{234}Th in

seawater. Distribution coefficients, K_d, ranges from $10^{5.4}$~$10^{6.4}$ ml/g, in the same range of previous studies in the vicinity (Hung and Wei, 1992; Wei et al., 1994) and other marginal sea (Baskaran et al., 1996).

According to the irreversible scavenging model the particle removal rate for ^{234}Th (i.e., P flux, Coale and Bruland, 1985) can be calculated by

$$P = \lambda (U - TTh) \qquad (1)$$

Where λ is the decay constant of ^{234}Th (0.0288 d^{-1}), U is ^{238}U activity (dpm/L), and TTh is total ^{234}Th activity (dpm/L). It should be noted that the P flux calculated from (1) has a rate unit, dpm/L/d, and P flux can be converted into a flux unit, dpm/m^2/d, by trapezoidal integration of P flux,

$$F_{Th} = \Sigma [(Pi + Pi\text{-}1) (Zi\text{-}Zi\text{-}1)/2] \qquad (2)$$

Where Pi is successive P flux calculated from discrete ^{234}Th data at depth Zi in the water column.

Vertical fluxes of ^{234}Th calculated from the irreversible model are shown in Figure 4. At station HT, ^{234}Th fluxes calculated from the two casts are remarkably similar and increases from ~150 dpm/m^2/d near surface to ~8200 dpm/m^2/d at 300 m. Th-234 fluxes also increase with depth at station B2 but with a lower increasing rate relative to depth than at station HT, ranging from 190 dpm/m^2/d near surface to 5800 dpm/m^2/d at 350 m. Th-234 fluxes estimated at nearby locations (OR1-306A and OR1-368, Figure 1) fall in the same range of this study (Hung and Wei, 1992 , Wei et al., 1994). The core top samples collected from a station (water depth = 46 m) to the north of this study (OR1-438, Figure 1) gave an inventory of excess ^{234}Th of 2.36×10^5 dpm/m^2 (Chu, 1996). Consequently, a ^{234}Th flux of 6790 dpm/m^2/d, scavenged from the overlying water column to the bottom sediments, is needed to support the excess ^{234}Th inventory.
Different from the observation that floating traps may not provide accurate vertical particle fluxes in some oceanic regimes (Bueseller, 1991), our previous results from floating trap showed excellent agreement between vertical ^{234}Th fluxes measured by traps and from ^{234}Th/^{238}U disequilibria in the study area (Wei et al., 1994).

Considering the resemblance of turn over time between ^{234}Th and POC in the euphotic zone, Eppley (1989) suggested that POC flux can be calculated from the

quotient of POC inventory and the residence time of ^{234}Th in the euphotic zone. The notation of treating ^{234}Th as a carbon tracer in the euphotic zone has been implemented in the framework of JGOFS (Joint Global Ocean Flux Studies) program. Assuming the difference of the inventories between ^{238}U and total ^{234}Th (dissolved + particulate) gives an accurate flux of settling particles from the euphotic zone, POC fluxes can be calculated by multiplying POC/^{234}Th ratio in suspended or settling particles to F_{Th} (Buesseler et al. 1995; Bacon et al., 1996; Murray et al., 1996) and the export carbon fluxes can then be calculated, i.e.,

$$F_{POC} = F_{Th}\ (POC/^{234}Th) \qquad (3)$$

where POC/^{234}Th is the average POC/^{234}Th ratio at depth Zi and Zi-1.
However, it should be noted that export flux of particulate organic carbon calculated by this model is highly dependent on the POC/^{234}Th ratio used and the ratio varies both temporally and spatially (Buesseler, 1998). Hence, in order to obtain an accurate estimation of POC export flux, it is required to simultaneously measure ^{234}Th /^{238}U deficiency in seawater and POC/^{234}Th ratio in particulates.

Vertical profiles of particulate ^{234}Th (PTh), POC, chlorophyll, and POC/PTh ratio at stations HT and B2 are shown in Figure 5 and 6, respectively. As seen in Figures 5 and 6, little similarity is shared among the vertical structures of PTh, POC, and chlorophyll. Representing a small pool of ^{234}Th in seawater, PTh ranges between 0.1 and 0.6 dpm/L and shows neither vertical nor horizontal trend. On the contrary, POC distribution is characterized by elevated concentration in the surface layer at both stations and in the bottom layer of station HT. Higher POC concentration at station HT than station B2 and elevated concentration in the bottom layer of station HT indicate the terrestrial influence to the nearshore station. Terrestrial influence can also be supported by the incoherent vertical structures between POC and chlorophyll distributions. Neither maximal POC nor maximal PTh concentration was observed at the subsurface chlorophyll maximum, which is typically found in the region. In consistent with the observation in Northern Iberian Margin off Spain (Hall et al., 2000), most of POC/^{234}Th ratio in the suspended particles fall in the range of 5 and 25 μmol/dpm, with considerable variability in the surface layer. The suspended particles residing in the upper water tend to have a

higher POC/^{234}Th ratio due to POC enrichment in the particulates.

The discussion on which size of particles is the most appropriate pool to be used for calculating F_{POC} has been raised (e.g., Bacon et al., 1996; Santschi et al., 1999; Rutgers van der Loeff, 1997) and consensus has not yet been reached on this issue. The nature of our dataset restricts us to use POC/^{234}Th ratio in total suspended particles larger than 0.45 μm for POC flux calculation. The results may be treated as an upper limit of the true export flux of POC if higher POC/^{234}Th were found in finer particles than large particles (Bacon et al., 1996; Santschi et al., 1999; Hall et al., 2000).

Vertical profiles of POC fluxes calculated from (3) were shown in Fig. 7. Except in the subsurface layer, the calculated POC fluxes are quite similar between the two stations, with the values of 10 and 70 mM/m^2/d in the surface and bottom layers, respectively. Overall, export flux of POC obtained in this study is an order of magnitude higher than oligotrophic open ocean (Buesseler et al. 1995; Bacon et al., 1996; Murray et al., 1996), implying the importance of coastal ocean in sequestering carbon via particle settling from upper water. Such a high POC fluxes were also found in the continental margin (Hall et al., 1999) and in eutrophic open ocean (Rutgers van der Loeff, 1997). In the subsurface layer, much higher POC fluxes were found at station HT than station B2. It is noted that POC fluxes in the subsurface layer of station HT show a large variability with a much higher POC flux at 2 p.m. and lower POC flux at 8 p.m., indicating a diurnal variability of carbon flux. From cast #26 obtained at 2 p.m. at station HT, POC flux reaches to 145 mM/m^2/d at 75 m then decreases dramatically to 25 mM/m^2/d at 150 m, implying that more than 80% of particulate organic carbon was decomposed and/or respired on the path of settling through the layer between 75 and 150 m. Repeated sampling of hydrographic parameters enables us to observe the temporal variability in the water column of station HT. Although not shown here, a layer of maximal nitrite concentration (> 0.5 μM), centered at 80 m, started to emerge from midnight of 29 March till 6 p.m. of 30 March. Milder subsurface maximum of POC flux from the profile of cast #8, obtained 2 hours prior to the emergence of nitrite maximum, may be related to the absence of the nitrite maximum during this time of the day. Lower production rate of POC in the surface layer and slower decomposition/respiration rate in the subsurface layer during nighttime is

hypothesized as the cause of the difference of POC flux between the two casts.

Conclusions

The degree of $^{234}Th/^{238}U$ disequilibria in the water column of two stations off southwestern Taiwan was investigated and used to estimate export production. Enhanced scavenging and removal processes were found in the coastal regime, which results in large ^{234}Th deficiency throughout the whole water column. A daily total of 5800~8200 dpm/m^2 ^{234}Th were stripped from the water column to the bottom sediments. POC fluxes, a proxy of export production, was calculated by multiplying ^{234}Th fluxes to the ratio of POC and ^{234}Th in the suspended particles. An average POC flux of 70 mM/m^2/d settling from the water column into sediment of the study area was estimated.

Acknowledgements

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Figure captions

Figure 1. Sampling stations (HT, B2) of OR1-545 occupied during 28~31 March 1999 off southwest Taiwan. Bottom depths of station HT and B2 are 347m and 395 m, respectively. Locations of sampling station from previous cruises were also shown.

Figure 2. (a) Vertical profiles of the concentration of total suspended matter, total, dissolved and particulate ^{234}Th collected from cast #8 at station HT. The vertical dashed line at 2.45 dpm/L represents ^{238}U activity calculated from S-^{238}U relationship. (b) Same as (a) except from cast #26.

Figure 3. Vertical profiles of the concentration of total suspended matter, total, dissolved and particulate ^{234}Th at station B2. The vertical dashed line at 2.45 dpm/L represents ^{238}U activity calculated from S- ^{238}U relationship.

Figure 4. Vertical profiles of calculated ^{234}Th fluxes from the cast #8 (●) and cast #26 (○) of station HT and station B2(□).

Figure 5.Vertical profiles of (a) particulate ^{234}Th, (b) particulate organic carbon, (c) chlorophyll concentration, and (d) ratio of organic carbon and ^{234}Th in suspended particulates of station HT. Samples taken from casts #8 and #26 are shown in open and closed circles, respectively.

Figure 6. Vertical profiles of (a) particulate ^{234}Th, (b) particulate organic carbon, (c) chlorophyll concentration, and (d) ratio of organic carbon and ^{234}Th in suspended particulates of station B2.

Figure 7. Vertical profiles of calculated fluxes of particulate organic carbon from the cast #8 (●) and cast #26 (○) of station HT and station B2(□).

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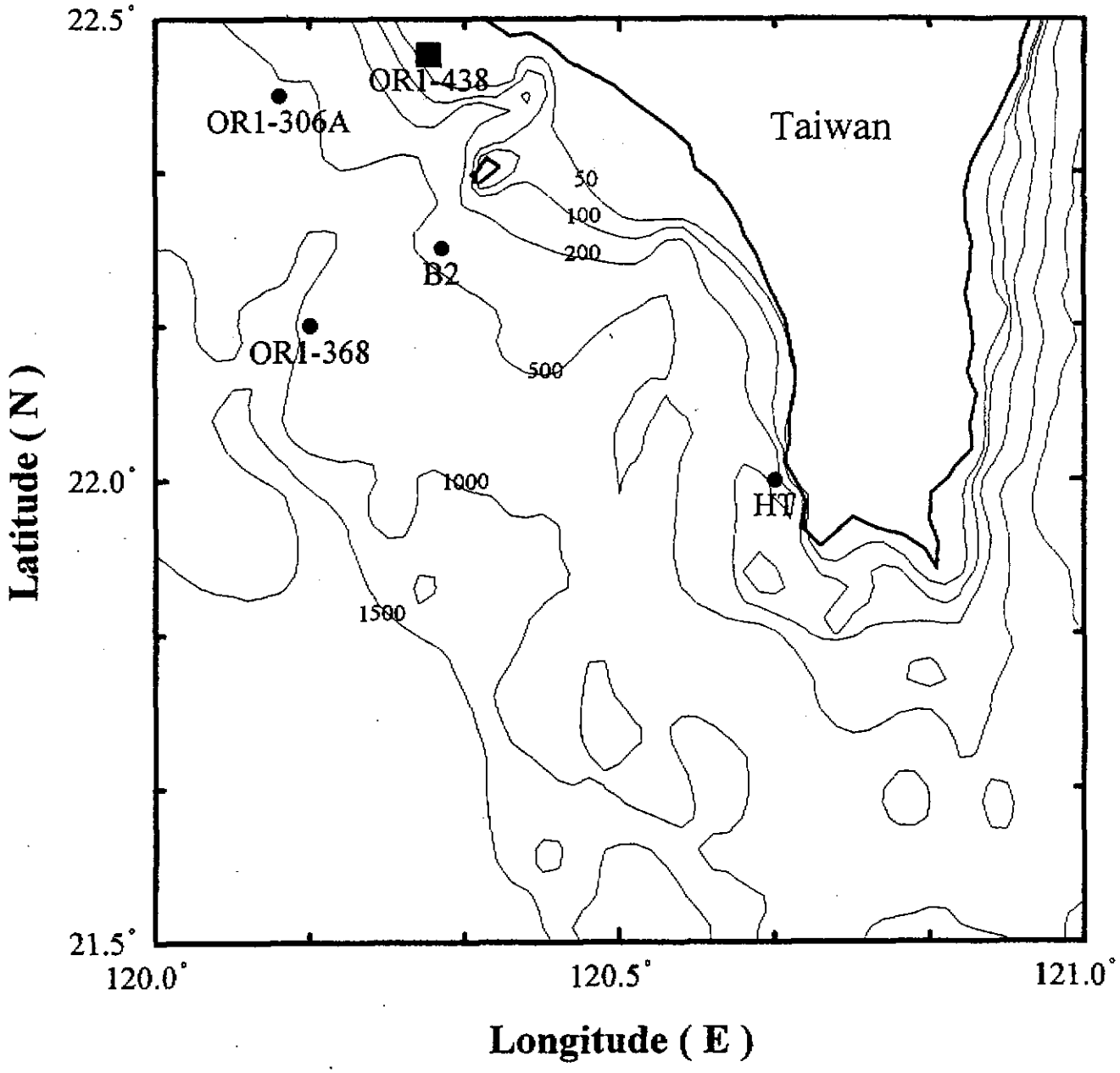
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B2
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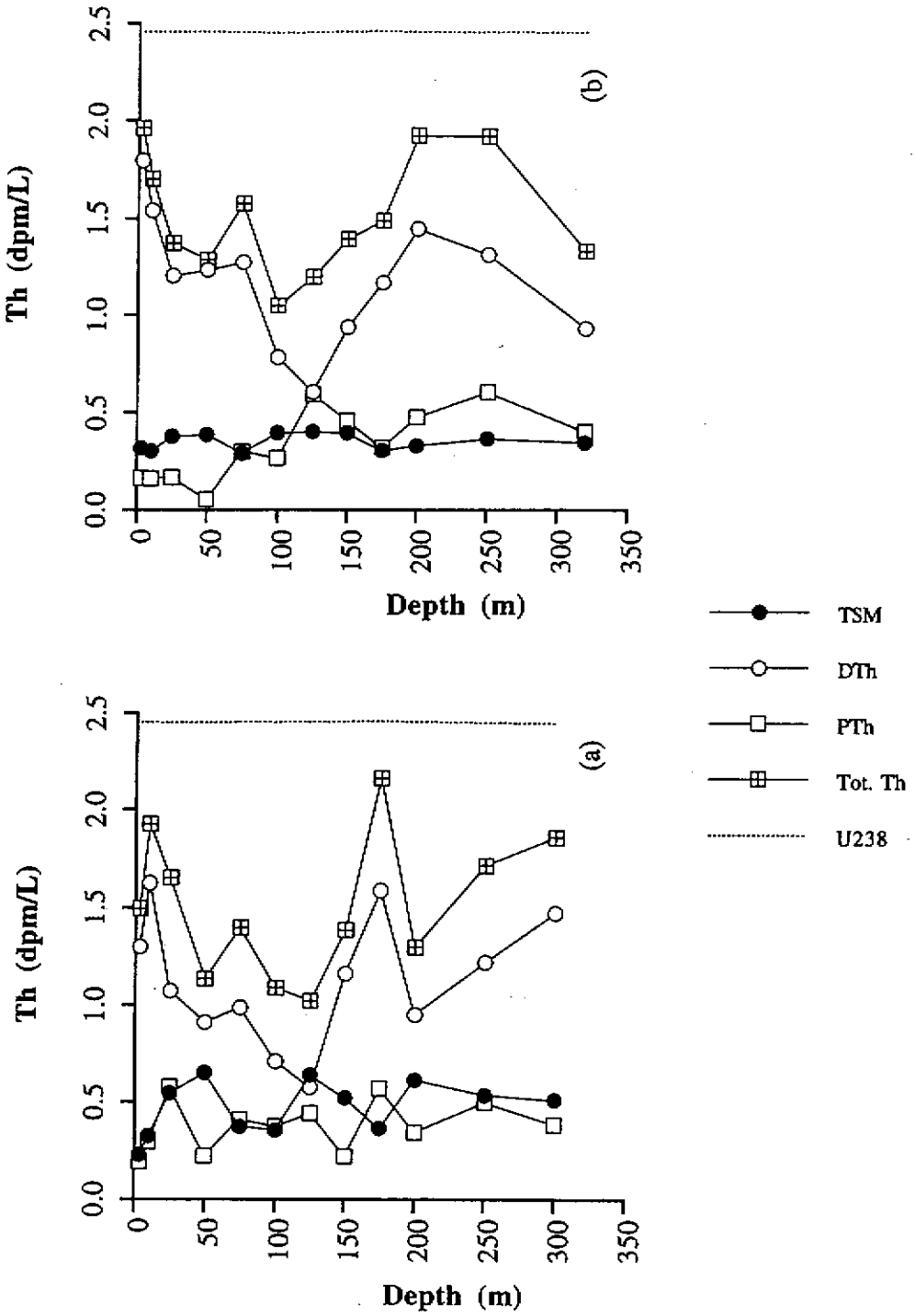
(a)
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Th (dpm/L)
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TSM
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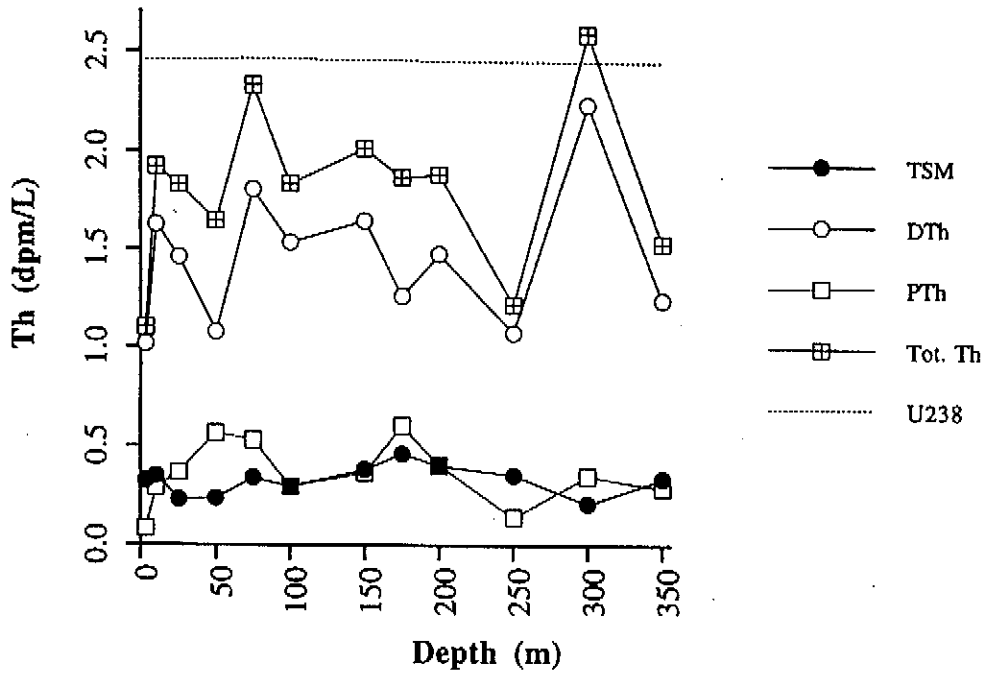
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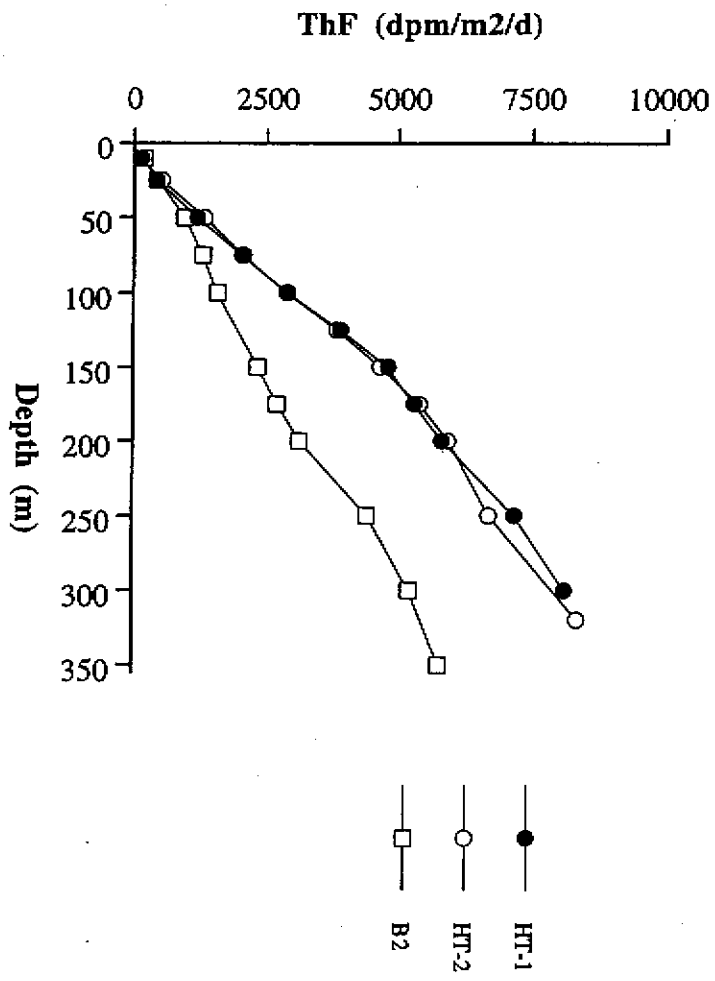
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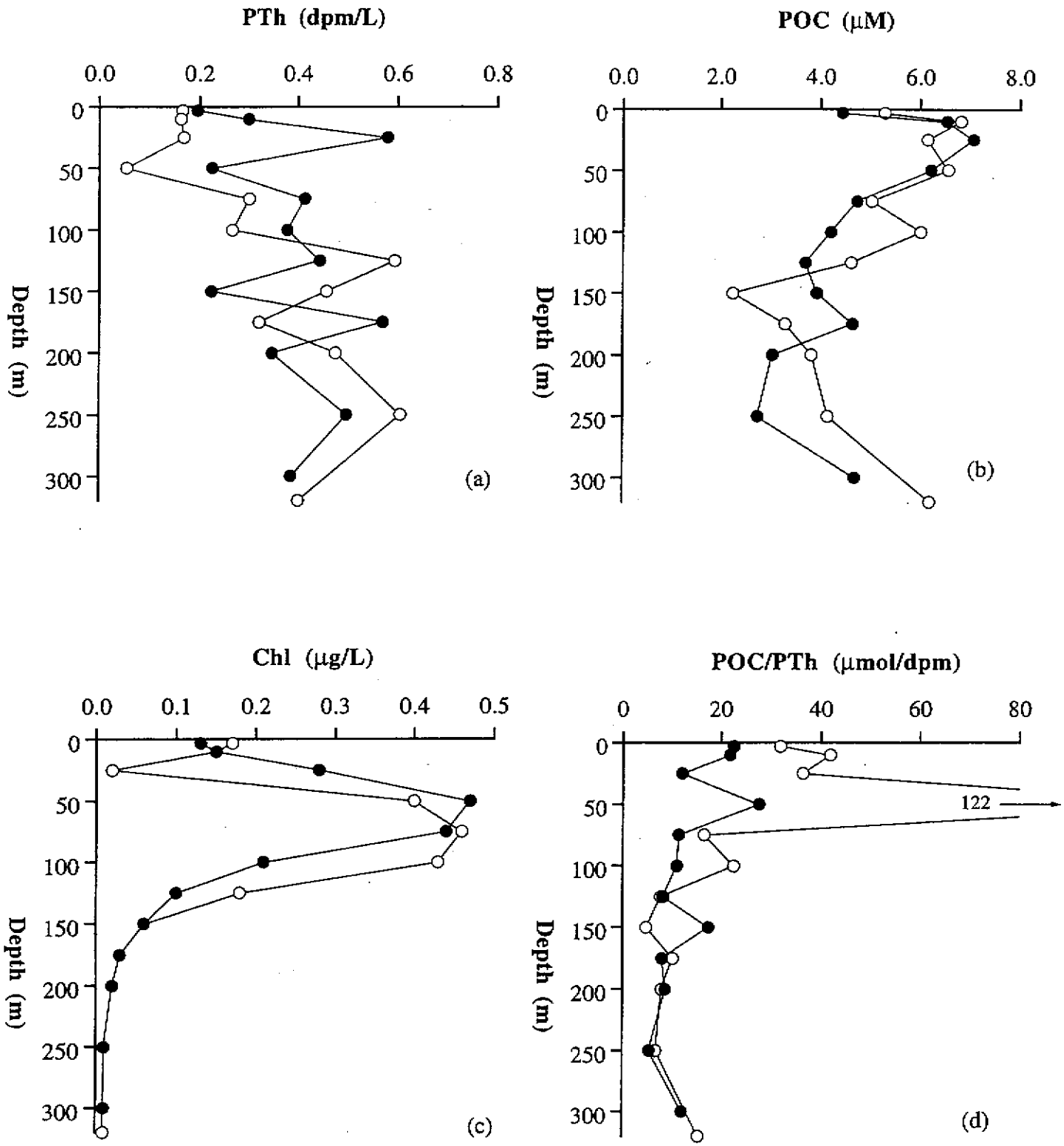
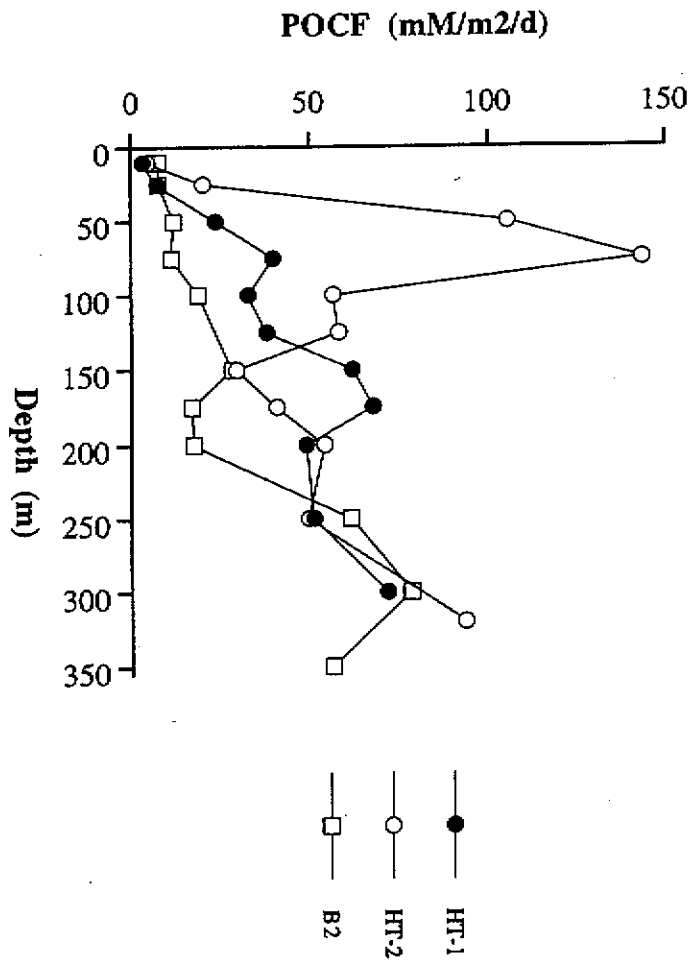
200
250
300
350
B2
HT-2
HT-1


PTh (dpm/L)
0.0
0.2
0.4
0.6
0.8
Depth (m)
0
50
100
150
200
250
300
(a)
POC (μM)
0.0
2.0
4.0
6.0
8.0
Depth (m)
(b)
Chl (μg/L)
0.0
0.1
0.2
0.3
0.4
0.5
Depth (m)
(c)
POC/PTh (μmol/dpm)
0
20
40
60
80
122
Depth (m)
(d)


HT-1
HT-2
B2
0
50
100
150
200
250
300
350
Depth (m)
0
50
100
150
POCF (mM/m2/d)