We are grateful to the Associate Editor, PA Meyers and an anonymous reviewer for their constructive feedbacks and insightful comments on our manuscript entitled "Biogeochemical characteristics of suspended particulates at deep chlorophyll maximum layers in the southern East China Sea" (**MS. Ref. No.: bg-2017-290-R1**).

Referee 1 (Prof. P. A. Meyers)
Liu and colleagues have nicely revised their interesting paper that describes the results of their study of the organic carbon and nitrogen contents in suspended particles collected around deep chlorophyll maximum layers in the East China Sea. The English has been much improved, and the whole paper is now easier to read and appreciate. I suggest a few minor additional refinements that might be considered by the authors before the contribution is ready to be published in Biogeosciences:

Reply: Thank you.

Referee 1: Title – change to read "'….suspended particulate matter in deep……"

Reply (Page 1, Lines 1-2): Changed.

Referee 1: Page 1, line 23 – replace "particulates" with "particles" (particulate is an adjective, not a noun)

Reply (Page 1, Line 19): Replaced.

Referee 1: Page 2, line 2 – change to read "additional radiocarbon and biomarker data are needed to re-evaluate"

Reply (Page 2, Lines 4-6): This sentence has been changed as follows:

Nonetheless, additional radiocarbon and biomarker data are needed to re-evaluate whether or not the POM around the DCM water depths is influenced by terrestrial OM in the river-dominated East China Sea.

Referee 1: Page 8, line 27 – change to read "near northeast Taiwan"

Reply (Page 9, Line 19): Changed.

Referee 1: Page 9, line 27 – change to read "influenced by the bottom"

Reply (Page 10, Lines 18-22): By combining suggestions of Referees 1 and 4, this sentence has been changed as follows:

Interestingly, the influence of CDW was constrained to the upper 10 m in five coastal stations, whereas TWCW influenced the upper 30 m and covered three quarters of the study region,

with KSSW largely influencing the bottom water across the entire study region (Figs. 2, 6a and 7).

Referee 1: Page 9, line 30 – change to read "transported northeastward of the"

Reply (Page 10, Line 26): Changed.

Referee 1: Page 10, line 23 – change to read "to more intense biological"

Reply (Page 11, Line 19): Changed.

Referee 1: Page 10, line 29 – change to read "ratio to more than that of"

Reply (Page 11, Line 25): Corrected.

Referee 1: Page 14, line 2 – change to read "biomass with increasing"

Reply (Page 14, Line 32): Changed.

Referee 1: Page 15, line 18 – change to read "may not result from the high degree of nitrate utilization, but instead from the"

Reply (Page 16, Lines 15-17): This sentence has been changed as follows:

There is another possibility that high $\delta^{15}N_{PN}$ (DH7-8: 6.7 ‰, DH7-9: 7.8 ‰) in the DCM layer, off northeast Taiwan (Fig. 5), may not result from the high degree of nitrate utilization, but instead from the incorporation of inorganic nitrogen (mainly NH_4^+) in the POM.

Referee 1: Page 17, line 10 – change "accounted" to "accounting"

Reply (Page 18, Line 12): Corrected.

Referee 1: Page 17, line 13 – replace "but" with "which"

Reply (Page 18, Line 13): Replaced.

Referee 1: Page 17, line 15 – change "low" to "lower"

Reply (Page 18, Line 17): Changed.

Thank you very much.

Anonymous Referee #4

General overview:

Liu et al. present isotopic compositions and concentrations of particulate organic carbon and particulate nitrogen (POC and PON, respectively) in samples collected from the deep chlorophyll maximum (DCM) in the south East China Sea in summer 2013. They combine these data sets with temperature, salinity, turbidity and calibrated chlorophyll fluorescence data to determine the sources of particulate organic matter in the DCM and what factors govern isotopic dynamics of POC and PON in the DCM. The authors attribute variation in $\delta^{13}C_{POC}$ to be governed by changes in primary productivity and community composition and variation in $\delta^{15}N_{PN}$ to be governed by changes in uptake of dissolved inorganic nitrogen (i.e. NH_4^+ or NO_3^-) and source (with a link to water masses). The handling and interpretation of isotopic data is good, however with a lack of certain auxiliary data sets (e.g. dissolved inorganic nutrients, community composition), and full methodological detail in the determination of others (chlorophyll), it is hard to fully critique their interpretation of the data and their conclusions. Following revision, this study could contribute to our understanding of organic matter cycling and production within shelf seas.

My primary concerns with the manuscript are as following:

- A lack of information in the methods relating to chlorophyll (sample treatment, analysis and calibration of the fluorescence sensor).
- Interpretation of $\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$ is sometimes highly speculative and in some cases data to support certain arguments are not provided (e.g. dissolved inorganic nutrient data).
- Grammar and sentence structure need improvement in some sections.

Specific points:

Referee 4: Introduction

P2, L8: Suggest the following grammatical changes to paragraph one of the introduction. Note I also suggest inserting a reference to support the statement of typical isotopic values for end members.

"Stable isotopes of organic carbon and nitrogen ($\delta^{13}C$, $\delta^{15}N$) and molar carbon to nitrogen (C/N) ratios are natural tracers frequently used to identify the source and fate of terrestrial organic matter (OM) in estuarine and marine environments (Meyers, 1994; Hedges et al., 1997; Goñi et al., 2014; Selvaraj et al., 2015). This approach is based on $\delta^{13}C$, $\delta^{15}N$ and C/N ratios being significantly different between different end-members (e.g. terrestrial and marine), and the assumption that only conservative physical mixing of bulk properties occur in these marginal settings (Thornton and McManus, 1994; Hedges et al., 1986). Quantifying the relative

contribution of end-members using mass balance models thus requires known and constant elemental and isotopic values of end-members and major sources of OM in the study region (e.g., Goñi et al., 2003). Therefore, application of mixing models for the discrimination of OM sources discrimination requires clearly identified representative values for local OM sources. However, often end-member values of $\delta^{13}C$, $\delta^{15}N$ and molar C/N ratios are represented by 'typical' numbers, such as ca. –20 ‰ and –27 ‰ for $\delta^{13}C$ of marine phytoplankton and terrestrial plants [INSERT SUPPORTING REFERENCE], respectively, but without measuring discrete end-member values in real, local or regional OM source materials. For example, a number of earlier studies failed to measure isotopic values of marine phytoplankton despite using end-member mixing models to distinguish marine versus terrestrial OM in surface sediments (e.g., Kao et al., 2003; Wu et al., 2013), or distinguish marine phytoplankton values from bulk surface particulate organic matter (POM) values (e.g., Zhang et al., 2007), or allochthonous POM (e.g., Hale et al., 2012). As POM in estuarine and marine systems is mostly derived from primary production, the stable isotope values ($\delta^{13}C$, $\delta^{15}N$) and molar C/N ratios of POM are largely representative of phytoplankton biomass (Gearing et al., 1984). Therefore, since phytoplankton are the main primary producers of marine OM, the elemental and isotopic compositions of phytoplankton should be considered while studying the dynamics of POM in the marine water column."

Reply (Page 2, Lines 10-35): This paragraph has been changed appropriately as follows:.

Stable isotopes of organic carbon and nitrogen ($\delta^{13}C$, $\delta^{15}N$) and molar carbon to nitrogen (C/N) ratios are natural tracers frequently used to identify the source and fate of terrestrial organic matter (OM) in the estuarine and marine environments (Meyers, 1994; Hedges et al., 1997; Goñi et al., 2014; Selvaraj et al., 2015). This approach is based on the significant difference in $\delta^{13}C$, $\delta^{15}N$ and C/N ratios between different end-members (e.g., terrestrial and marine), and the assumption that only a physical mixing of OM from compositionally distinct end-members occurs in these marginal settings (Thornton and McManus, 1994; Hedges et al., 1986). Quantifying the relative contributions of end-members using mass balance models thus requires known and constant elemental and isotopic values of end-members and major sources of OM in the study region (e.g., Goñi et al., 2003). Therefore, application of mixing models for the discrimination of OM sources requires clearly identified representative values for local OM sources. However, in most cases, end-member values of $\delta^{13}C$, $\delta^{15}N$ and molar C/N ratios are represented by 'typical' numbers, such as ca. –20 ‰ and –27 ‰ for $\delta^{13}C$ of marine phytoplankton and terrestrial plants (Kandasamy and Nagender Nath 2016 and references therein), respectively, but without measuring discrete end-member values in real, local or regional OM source materials. For example, a number of earlier studies failed to measure isotopic values of marine phytoplankton despite using end-member mixing models to distinguish marine versus terrestrial OM in surface sediments (e.g., Kao et al., 2003; Wu et al., 2013), or these numbers simply represented by values of particulate organic matter (POM) in surface waters in the studied system (e.g., Zhang et al., 2007) or elsewhere from other ocean basins (e.g., Hale et al., 2012). It is known that stable isotopes ($\delta^{13}C$, $\delta^{15}N$) and molar C/N ratios of POM in estuarine and marine areas are representative of primary production-derived OM when POM are mostly derived from phytoplankton biomass (Gearing et al., 1984). Since

phytoplankton are the main primary producer of marine OM, the elemental and isotopic compositions of phytoplankton should therefore be considered while studying the dynamics of POM in the marine water column.

Referee 4: P2, L30 – L31: Please delete "and phytoplankton carbon". The C: Chl ratio of phytoplankton can vary with species, depth and nutrient status and so conversion of Chl a concentration to C concentration can involve significant errors.

Reply (Page 3, Lines 1-2): Deleted.

Referee 4: P2, L30 – P3, L10: Subsurface or deep chlorophyll maxima (SCM and DCM, respectively) form within the thermocline in shelf sea systems. To my knowledge the formation of SCM in shelf sea systems is largely linked to turbulence, diapycnal nutrient fluxes and light acclimation. I recommend the following papers for further detail and references therein:

Sharples et al. (2001) Phytoplankton distribution and survival in the thermocline, L&O, DOI: 10.4319/lo.2001.46.3.0486
Moore et al. (2006) Phytoplankton photoacclimation and photoadaptation in response to environmental gradients in a shelf sea, L&O DOI: 10.4319/lo.2006.51.2.0936
Hickman et al. (2012) Primary production and nitrate uptake within the seasonal thermocline of a stratified shelf sea, MEPS, DOI: 10.3354/meps09836
Williams et al. (2013) Wind-driven nutrient pulses to the subsurface chlorophyll maximum in seasonally stratified shelf seas, GRL, DOI: 10.1002/2013GL058171

Reply (Page 3, Lines 6-12): Thanks for providing these important references on the DCM formation in the shelf seas. Based on some of these references, we revised the text as follows:

The formation of maximum chlorophyll concentration at the DCM layer has been explained by several mechanisms: the differential zooplankton grazing with depths (Riley et al., 1949; Lorenzen, 1967), adaption of phytoplankton to light intensities or to increased concentration of nutrients (Nielsen and Hansen, 1959; Gieskes et al., 1978; Hickman et al., 2012), chlorophyll accumulation by sinking detritus of phytoplankton (Gieskes et al., 1978; Karlson et al., 1996), decomposition of chlorophyll by light (Nielsen and Hansen, 1959), and wind-driven nitrate supply and nitrate uptake in seasonally-stratified shelf seas (Hickman et al., 2012; Williams et al., 2013).

Referee 4: P3, L13: replace "carbon" with "C", note: be consistent with abbreviations, once an abbreviation is defined e.g. carbon (C), or East China Sea (ECS) be sure to use the abbreviation from then on.

Reply (Page 3, Line 22): Replaced.

Referee 4: P3, L14: Suggest the following grammatical changes: "Nutrient-rich freshwater inputs in turn stimulate water column productivity in coastal water compared to the open ocean.

Annual primary production over the entire shelf of the East China Sea is high relative to other marginal seas and was estimated to be ….”

Reply (Page 3, Lines 22-25): Corrected, as suggested.

Referee 4: P3, L18: Replace “nutrients” with “nutrient”

Reply (Page 3, Line 27): Replaced.

Referee 4: P3, L19: Insert “composition” after “phytoplankton species”

Reply (Page 3, Line 27): Inserted.

Referee 4: P3, L20: Replace “constrained in” with “determined for”

Reply (Page 3, Line 29): Replaced.

Referee 4: P3, L22: Suggest the following grammatical changes: “Nonetheless, studies on elemental ratios and stable isotopic compositions of POM in DCM layers in the East Chin continental shelf sea, especially transfers between the Yangtze and Okinawa Trough are poorly studied (Chen et al., 2017).”

Reply: We disagree with the Referee’s view here because there are three pathways of material transfer between the Yangtze and Okinawa Trough. Therefore, we prefer to keep our original text here to specify “along the indirect pathway of the Yangtze-derived terrestrial material to the Okinawa Trough”. Thank you.

Referee 4: P3, L24: Suggest the following grammatical changes: “A recent study in the north East China Sea investigated elemental and isotopic compositions of POM in the surface, DCM and bottom layer on both seasonal and inter-annual timescales (Gao et al., 2014), however there was minimal attention given to biogeochemical processes associated with the DCM.”

Reply (Page 4, Line 4 and Page 5, Lines 1-4): As suggested, the sentence has been modified as follows:
A recent study in the northern East China Sea investigated elemental and isotopic compositions of POM in the surface, DCM and bottom layers on both seasonal and inter-annual timescales (Gao et al., 2014); however, there was minimal attention given to biogeochemical processes associated with the DCM.

Referee 4: P3, L28: Replace “around” with “at”, delete “layer”, insert “South” before “East China Sea”.

Reply: Some samples investigated in this study fall contiguous to the DCM layer and therefore we prefer to keep our original text here. Thank you.

Referee 4: P3, L29: Replace "comprehend" with "determine".

Reply: We think here "comprehend" is better than "determine", since OM sources' identification in this study was based on multi-data analyses.

Referee 4: P4, L1: Please insert a reference to support this statement.

Reply (Page 4, Line 11): Inserted.

Referee 4: P4, L2: Suggest the following grammatical changes: "The ECS shelf is wide (>500 km), but relatively shallow (<130 m) with an average water depth of 60 m (INSERT REFERENCE)".

Reply (Page 4, Lines 12-13): This sentence has been changed as follows:

The ECS shelf is wide (>500 km), but relatively shallow (<130 m) with an average water depth of 60 m (Gong et al., 2003; Liu et al., 2006).

Referee 4: P4, L3: Suggest the following restructuring: "With a catchment area of more than 1.94 x 10^6 km^2 (Lui et al., 2007) resulting in an annual freshwater discharge of 900 km^3 yr^{-1}, the fifth largest in the World, and a sediment discharge of 470 Mt yr^{-1}, the fourth largest in the World (Milliman and Farnsworth, 2011), the Yangtze River is the main source of freshwater and sediment in the ECS."

Reply: As given by the referee, this sentence is too long! In general, the scientific literature prefers shorter and meaningful sentences. We therefore keep the original text here without any change.

Referee 4: P4, L12: Replace "middle" with "central". Is "north" intentionally repeated?

Reply (Page 4, Lines 22-23): Corrected.

Referee 4: P4, L13: Suggest the following: "The Changjiang Diluted Water (CDW) is a mixture of Yangtze River freshwater and ECS shelf water and is characterised by…".

Reply (Page 4, Lines 23-25): Corrected.

Referee 4: P4, L16: Replace "it has been believed" with "it is thought that", perhaps replace "source" with "component".

Reply (Page 4, Lines 26-27): Replaced.

Referee 4: P4, L16: Suggesting the following: "In winter, the CDW flows southwards along the coastline of a mainland China as a narrow jet (Chen, 2008; Han et al., 2013) and in summer

spreads to the northeast (Isobe et al., 2004). These changes are driven by the East Asian monsoon, which constitutes a strong northeast monsoon in winter and weaker southwest monsoon in summer."

Reply: Since we are unhappy about the referee's mere shifting of sentences in the name of grammar and restructuring, we prefer to keep the original text as such here.

Referee 4: P4, L19: Insert "The" before "Taiwan Warm Current".

Reply (Page 4, Line 30): Inserted.

Referee 4: P4, L20: Delete "the" before "intruding".

Reply (Page 4, Line 32): Deleted.

Referee 4: P4, L21: Suggest: "In addition, Kuroshio Subsurface Water (KSSW) is upwelled in the northeast near Taiwan Island due to an abrupt change in seafloor topography at the ECS outer shelf…"

Reply (Page 4, Lines 33-35): This sentence has been changed appropriately as follows:

In addition, Kuroshio Subsurface Water (KSSW) is upwelled in the northeast off Taiwan Island due to an abrupt change in seafloor topography at the ECS outer shelf

Referee 4: P4, L24: Perhaps "oxygen-under saturated". Delete "but".

Reply: Sorry! To the best our knowledge, oxygen-unsaturated is more scientific and widely used while dealing biogeochemical cycles/processes. Please refer to the definition of oxygen saturation – *a ratio of the concentration of dissolved oxygen (O_2) in the water to the maximum amount of oxygen that will dissolve in that water.* Deleted (**Page 5, Line 1**).

Referee 4: P4, L25: Replace "East China Sea" with "ECS".

Reply (Page 5, Line 2): Replaced.

Referee 4: P4, L27: Suggest: "Furthermore, Kuroshio water accounts for up to 90 % of shelf waters in the ECS…"

Reply (Page 5, Lines 5): Corrected.

Referee 4: P4, L30: Suggest: "Primary production in the ECS is nutrient-limited in summer and light-limited in winter (Chen et al., 2001; Chen and Chen 2003), with production being higher in summer. In 2008, annual primary production rates showed distinct spatial variation, with rates in the north-western ECS (155 g C m^{-2} y^{-1}) being higher than those of the south-eastern ECS

(144 g C m^{-2} y^{-1}) and the overall average for the ECS (145 g C m^{-2} y^{-1}) (Gong et al., 2003). However, primary production rates have decreased by 86 % between 2008 and 2003, due to installation of a number of reservoirs within the Yangtze River drainage basin (Gong et al., 2006).

Reply: Since we are unhappy about the referee's version, we prefer to keep the original text as such here because that is more simple and understandable than the referee's version.

Referee 4: Material and methods

P5, L4: I think it is usually "Materials and methods"

Reply (Page 5, Line 14): Corrected.

Referee 4: P5, L4: "Water samples were collected at 36 stations along seven transects in the ECS during the Science 3 cruise in summer (June 22 – July 21) 2013 (Fig. 1). Water samples were collected from X? DCM depths (10 -130 m; Table 1) at each station using ?L Niskin bottles mounted on a sampling rosette. A Seabird Conductivity-Temperature-Depth (CTD, SBE911+) sensor fitted with a calibrated? Seapoint chlorophyll fluorometer was mounted on the rosette to record the physical properties of the water column and the depth of the DCM, respectively."

Reply: These are standard ways of collecting and analyzing samples and measuring onboard parameters and researchers throughout the world are following these standardized procedures. Details given in the original version are clear and concise enough to the readers.

Referee 4: P5, L24: If the filters were freeze dried, what is the rationale behind then drying them again at 50 °C for 48h?

Reply: This is the standard way to eliminate the influence of temperature and humidity on the determination of SPM weight.

Referee 4: P5, L25: "counterpart" is not the right word here, it implies two separate filters were weighed and their difference was considered to be the SPM weight.

Reply (Page 6, Line 2): "its counterpart" was changed as "the same filter".

Referee 4: P5, L28: If you randomly selected samples for Chl a analysis did you store all filters in the dark? Did the pigments survive the freeze-drying plus 48h drying at 50 °C? How did you prepare your standards? Did you take standards through the full drying/extraction process to check recoveries? How long was the acetone extraction? Further detail is needed here to convince the reader that chlorophyll concentration data are reliable.

Reply: All filters were wrapped in aluminum foil, which protected filters from light and stored at

–20 °C freezer onboard immediately after filtration. We used the monochromatic method with acidification to determine Chl a concentration by UV-Vis spectrophotometer and samples were measured against 90% acetone as blank.

This paragraph has been changed as follows (**Page 6, Lines 1-12**):
In the laboratory, filters with suspended particles were freeze-dried and then dried in an oven at 50 ºC for 48 h. The weight difference between the dried filter and the same filter before the filtration was used to calculate the weight of SPM. Five SPM samples (DH1-2, DH2-1, DH3-1, DH7-1 and DH7-7; Fig. S1) from water depths ranging between 20 m and 50 m were randomly selected for the measurement of chlorophyll a (Chl a) concentration. Chlorophyll a was extracted using 90% acetone and then determined spectrophotometrically according to Lorenzen (1967) and Aminot and Rey (2000). Briefly, the absorbance of sample extraction was measured at 665 nm and 750 nm against a 90% acetone blank before ($E665_o$, $E750_o$) and after ($E665_a$, $E750_a$) acidification with 1% HCl by the UV-Vis spectrophotometer (UV 1800, Shimadzu). Chl a concentration ($\mu g\ L^{-1}$) was calculated as: Chl a = 11.4 × 2.43 × (($E665_o$ − $E750_o$) − ($E665_a$ − $E750_a$)) × V_e /L × V_f, where V_e and V_f were the volumes of sample extraction and sea water filtered (mL), respectively, and L was the cuvette light-path (cm) (Aminot and Rey, 2000).

Referee 4: P5, L32: Please clarify if it was the half of the filter for POC analysis that was de-carbonated and whether the half for PN analysis was also subjected to decarbonation.

Reply: Please refer to our reply to Referee 2 comments in the previous round of review.

Referee 4: P6, L1: What diameter was the punch, you could say "transferred to tin capsules", which further analysis are you referring to here?

Reply: Please refer to our reply to Referee 2 comments in the previous round of review.

Referee 4: P6, L5: Suggest: "A range of working standards with compositional similarities to the samples were selected (bovine liver, glutamic acid, enriched alanine and nylon 6) and were calibrated against NIST Standard Reference Materials…"

Reply: The original text is better than the suggested by the referee. For instance, "A range was selected, not were"! We prefer to keep the original sentence here.

Referee 4: P6, L8: Replace "is" with "was".

Reply (Page 6, Line 25): Replaced.

Referee 4: P6, L14 – 23: You have raised this issue that your results may show some bias due to de-carbonation of the PN filters and you have provided evidence that your results are in line with those of Wu et al. (2003), who also de-carbonated their samples from this study region. It would perhaps be more useful to be able to quote similar values for this region obtained

without de-carbonation of PN filters to suggest that the bias resulting from the freezing and de-carbonation process did not significantly alter your results.

Overall, this paragraph could be improved by restructuring and better linking (or not linking) between sentences.

Reply: Thanks for your suggestion. However, this paragraph is included based on the suggestion of Referee 2 in the previous round of review. Thank you.

Referee 4: Results and interpretations

P6, L32: Suggest restructuring: "Water temperature in the upper 300-m varied from 15 to 30 °C, with distinct thermal stratification of the water column across the entire study area (Fig. 2)."

Reply (Page 7, Lines 15-17): Corrected.

Referee 4: P6, L33 – P7, L1: Suggest deleting this sentence as Fig. 2 does not show data from 850 m or 800 m.

Reply (Page 7, Line 23): We cite Table S1 where one can see temperatures of 850 m and 800 m.

Referee 4: P7, L2 – 3: The statement "showing a general decreasing trend from the inner to outer shelf in each transect" with respect to temperature in Fig. 2 does not appear to hold true for the top 4 of the 7 transects.

Reply: This is mainly because stations, DH1-1, DH2-1, DH3-1, CON02 and DH2-2 in the top 4 of 7 transects, that were influenced by SMW (see section 5.1 for more details).

Referee 4: P7, L5-13: The first and last sentences of this paragraph are repetitive, but quote different average salinity values. In addition, I do not feel that the sentence describing the "middle salinity" adds any useful information to the description of salinity. Therefore I suggest the following reorganisation or something similar: "The salinity distribution at depths of SPM sampling showed an increasing trend from the inner to outer shelf (Fig. 2), varying from 32.7 to 34.7 with an average salinity of 34.0 ± S.D. Low salinity water (<30) was observed in the upper 10-m at four of the coastal stations where water temperatures were <24°C (Fig. 2), suggesting that there was limited influence of the CDW plume in the study region. The highest salinities were observed at depth and off shelf (Fig. 2)."

Reply: The first sentence referred to salinity profile in the whole water column, while the last sentence referred to salinity at the sampling depths. Please check carefully. The description on "middle salinity" is necessary as it is related to the TWCW and SMW, and the same is discussed in section 5.1 with more details. In fact, the entire paragraph was restructured based on the correction of Referee 2 in the previous round of review. Referee 4 should refer to those corrections before providing more or less similar, but a distorting way of restructuring in the version already revised.

Referee 4: P7, L19: Replace "limited along the coast" with "restricted to coastal stations".

Reply (Page 8, Line 8): We feel that "stations were limited along the coast" is more appropriate than "stations were restricted to the coastal stations.

Referee 4: P7, L27-28: Suggest restructuring to "The highest Chl fluorescence concentration (18.0 µg L-1) was observed in surface waters at station DH3-1. All other values were less than 8.0 µg L-1 (Fig. 3)."

Reply (Page 8, Lines 12-14): Restructured.

Referee 4: P7, L29: "showed" not "show"

Reply (Page 8, Line 14): Corrected. However, when we describe our own results with figure and table citations, it is better to use the present tense than the past tense.

Referee 4: P7, L32: Replace "straddling around" with "across"

Reply (Page 8, Line 18): Replaced.

Referee 4: P7, L33: Delete the comma after depth

Reply (Page 9, Line 19): Deleted.

Referee 4: P8, L7: Delete "productivity".

Reply (Page 9, Line 26): Deleted.

Referee 4: P8, L20: "showed" not "shows"

Reply (Page 9, Line 7): Corrected.

Referee 4: P8, L21: "concentration" should be plural

Reply (Page 9, Line 8): Corrected.

Referee 4: P8, L24: "were" not "are"

Reply (Page 9, Line 14): Corrected.

Referee 4: P8, L25: Suggest restructuring this sentence as follows: "POC and PN concentrations were highest near the coast on the inner shelf (>90 µg L-1 and >21 µg L-1, respectively), and decreased gradually with distance offshore (Fig. 4)."

Reply (Page 9, Lines 12-15): Corrected.

Referee 4: P8, L27: Delete "nearby off" and insert "of" between "northeast" and "Taiwan"
Reply (Page 9, Line 16): Corrected.

Referee 4: P8, L28: Insert "by" between "varied" and "more".

Reply (Page 9, Line 17): Inserted.

Referee 4: P8, L29: Replace "of the entire ECS" with "throughout sampling". Please specify whether 5.6 ± 0.5 is the mean ± S.D. or mean ± 95% confidence interval.

Reply (Page 9, Line 18): Replaced. 5.6 ± 0.5 is the mean ± S.D. (see Table 1 for clarification).

Referee 4: P8, L34: "Consistent with the POC"

Reply (Page 9, Line 23): Corrected.

Referee 4: P9, L1-L3: Suggest the following restructuring: "The lowest $\delta^{13}C_{POC}$ values were observed northeast of Taiwan Island in the Okinawa Trough, whereas $\delta^{15}N_{PN}$ values in this region were higher than those of the surrounding area (Fig. 5)."

Reply (Page 9, Lines 25-28): Restructured as follows:

The lowest $\delta^{13}C_{POC}$ values (–25.8 ‰ and –25.2 ‰) were observed northeast of Taiwan Island in the Okinawa Trough, whereas $\delta^{15}N_{PN}$ values (6.73 ‰ and 7.78 ‰) in this region were higher than those of the surrounding area (Fig. 5).

Referee 4: P9, L4: "was" not "is"

Reply (Page 9, Line 28): Corrected.

Referee 4: Discussion
P9, L17: This sentence is repetitive, suggest deleting as the detail is already covered in the sentence before.

Reply (Page 10, Lines 6-7): Deleted.

Referee 4: P9, L21: Define SMW (if not already defined)

Reply (Page 10, Line 10): Defined.

Referee 4: P9, L22: Insert "at" between "except" and "these"

Reply (Page 10, Line 11): Inserted.

Referee 4: P9, L24: Suggest "…further delineated the area and water depths influenced by…"

Reply (Page 10, Lines 13-14): Corrected.

Referee 4: P9, L 25: Suggest "Interestingly, the influence of CDW was constrained to the upper 10 m in five coastal stations, whereas TWCW influenced the upper 30 m and covered three quarters of the study region, with KSSW largely influencing the bottom water across the entire study region (Fig. 2, 6a and 7)."

Reply (Page 10, Lines 14-18): Corrected.

Referee 4: P9, L30: Delete "to"

Reply (Page 10, Line 22): Deleted.

Referee 4: P11, POC/Chl a: I think it would perhaps be useful here to also note that both vertical gradients in community composition and photoacclimation can influence C/N and POC/Chla within subsurface chlorophyll maxima.
Moore et al. (2006) Phytoplankton photoacclimation and photoadaptation in response to environmental gradients in a shelf sea, L&O DOI: 10.4319/lo.2006.51.2.0936
Latasa et al. (2017) Distribution of phytoplankton groups within the deep chlorophyll maximum. L&O, DOI: 10.1002/lno.10452.

Reply: Thanks for your suggestions. We agree with the referee that light or photoacclimination also influences the phytoplankton groups/community distribution in the water column, and may further contribute to the variations of POC/Chl a ratio as well as carbon and nitrogen isotopes in DCM. More complex physical forcings are possible, but these are beyond the scope of the present study.

Referee 4: P12, L 10-14: Is the increasing trend of $\delta^{13}C$ evident in SPM and surface sediments from this study or from the literature? What C does it refer to? This is a little unclear.

Reply: The inner to outer shelf increasing trend of $\delta^{13}C$ in SPM and surface sediments was evident from the literature cited in **Page 13, Lines 4-5**. Carbon isotopic composition of particulate organic carbon - $\delta^{13}C_{POC}$.

Referee 4: P12, L34: This needs a supporting reference e.g. Burkhardt et al. (1999).

Reply (Page 13, Line 25): The following two references are included.

Falkowski, P. G.: Species variability in the fractionation of ^{13}C and ^{12}C by marine phytoplankton, J. Plank. Res., 13, 21–28, 1991.

Hinga, K. R., Arthur, M. A., Pilson, M. E. Q., and Whitaker, D.: Carbon isotope fractionation by marine phytoplankton in culture: The effects of CO_2 concentration, pH, temperature, and species, Global Biogeochem. Cy., 8, 91–102, 1994.

Referee 4: P14, L30 – P15, L6: How would local regeneration of N and regenerated production influence these isotopic values, in addition to the $\delta^{15}NO_3$ of source waters?

Reply: NO_3^- is considered as the dominant source of nitrogen in this region. In case if the regenerated N from OM was the nitrogen source and it is deficit relative to other nutrients (Si, P), then $\delta^{15}N_{PN}$ would show isotopic values similar to regenerated N.

Referee 4: P15, L17 – L33: I found this paragraph hard to follow and overly speculative.
"… may not be resulted from the high degree of nitrate utilisation, but the incorporation of inorganic nitrogen in the POM." Nitrate is inorganic nitrogen, do you mean that there is a higher proportion of particulate inorganic nitrogen relative to particulate organic nitrogen and that this could be driving the isotopic signal here?
"The low Chl fluorescence might be limited by the low temperature in this high nutrient low chlorophyll region." The depths where you sampled were quite warm and of similar temperature to one another, I think a temperature effect on Chl here is unlikely. These appear to be the deepest samples at ~100m. Is it possible that this signature of low Chl and low POM concentration is because you were sampling below the euphotic layer? This could possibly contribute to the isotopic signal at this station, as below the euphotic layer remineralisation and degradation of POM would exceed production. The $\delta^{13}C_{DIC}$ at depth may have been quite different to that in the euphotic layer. If dissolved inorganic nutrient data and PAR data are available and support your argument that it is a temperature effect then this should be included.
No NH_4^+ data are available, and no $\delta^{15}N_{NH4+}$ values have been previously published for this region, therefore I find the discussion linking the high $\delta^{15}N_{PN}$ to ammonium assimilation quite speculative.

Reply: We agree with the referee that some of our statements are speculative, but we explained why our speculation is valid in our reply to Referee 2 comments in the previous round of review. It is really unfortunate that Referee 4 here provided almost similar comments (~90%) what Referee 2 have raised in the previous round of review.

Referee 4: P16, L5: insert "respectively" after "Wu et al"

Reply (Page 16, Line 34): We used semicolon (not and!) in between these values, and therefore inserting "respectively" is not necessary here!

Referee 4: P16, L7: Suggest restructuring to this or similar: "Our results indicate that POM at the DCM was largely produced in situ and derived from phytoplankton biomass, with little terrestrial influence. The lack of terrestrial OM signals…"

Reply (Page 16, Line 35 and Page 17, Lines 1-2): Corrected as suggested.

Referee 4: P16, L10: "has been reduced"

Reply (Page 17, Line 5): Corrected.

Referee 4: P16, L12: "…reported that the particulate load discharged by…"

Reply (Page 17, Line 7): Corrected.

Referee 4: P16, L22: "Accompanying the decreasing…"

Reply (Page 17, Line 18): Corrected.

Referee 4: P17, L7: "…despite the study are being the best…"

Reply (Page 18, Line 3): Corrected.

Referee 4: P17, L9: As I understand it you did not measure primary production rates, you are inferring primary productivity from POC concentration. Therefore it may be more accurate here to say biomass rather than primary productivity. Again, to my knowledge you did not collect data on community composition and therefore I am concerned that this statement is overly speculative for your concluding remarks.

Reply: In our study, primary productivity was inferred from the good positive relationship between $\delta^{13}C$ and POC, instead from POC alone (see Page 12, Lines 24–L26 in the original version). In terms of phytoplankton biomass, Chl *a* may be an ideal parameters than POC.

Sufficient information on the community composition of ECS were referred in Page 13, Lines 1-15 of the original text, and that information is consistent with our $\delta^{13}C_{POC}$ results and concluding remarks of our study.

Referee 4: P17, L11: Again, linking changes in $\delta^{15}N_{PN}$ when no NH_4^+ or NO_3^- data or uptake data are available is perhaps overly speculative.

Reply: We agree with the referee's view and we therefore mentioned that the possibility of such mechanism "needs to be substantiated by the nutrient data in future studies" (**Page 18, Line 13**).

Referee 4: P17, L20: Please expand on the link between the DCM and the inner shelf mud-belt that accumulated during the Holocene.

Reply: In this sentence, we stress that $\delta^{13}C$ values of DCM of this study can provide an ideal marine organic carbon end-member to evaluate the carbon burial along the mud-belt during

the Holocene, i.e., form a basis for the long-term evaluation of organic carbon burial.

Biogeochemical characteristics of suspended ~~particulates~~ **particulate matter in** ~~at~~ deep chlorophyll maximum layers in the southern East China Sea

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Abstract. Continental shelves and marginal seas are key sites of particulate organic matter (POM) production, remineralization and sequestration, playing an important role in the global carbon cycle. Elemental and stable isotopic compositions of organic carbon and nitrogen are thus frequently used to characterize and distinguish POM and its sources in suspended ~~particulates~~ particles and surface sediments in the marginal seas. Here we
20 investigated suspended particulate matters (SPM) collected around deep chlorophyll maximum (DCM) layers in the southern East China Sea for particulate organic carbon and nitrogen (POC and PN) contents and their isotopic compositions ($\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$) to understand provenance and dynamics of POM. Hydrographic parameters (temperature, salinity and turbidity) indicated that the study area was weakly influenced by freshwater derived from the Yangtze River during summer 2013. Elemental and isotopic results showed a large variation in
25 $\delta^{13}C_{POC}$ (–25.8 to –18.2 ‰) and $\delta^{15}N_{PN}$ (3.8 to 8.0 ‰), but a narrow molar C/N ratio (4.1–6.3) and low POC/Chl *a* ratio (<200 g g^{-1}) in POM and indicated that the POM in DCM layers was newly produced by phytoplankton. In addition to temperature effects, the range and distribution of $\delta^{13}C_{POC}$ were controlled by variations in primary productivity and phytoplankton species composition; the former explained ~70% of the variability in $\delta^{13}C_{POC}$. However, the variation in $\delta^{15}N_{PN}$ was controlled by the nutrient status and $\delta^{15}N_{NO3^-}$ in seawater, as indicated by
30 similar spatial distribution between $\delta^{15}N_{PN}$ and the current pattern and water masses in the East China Sea; although interpretations of $\delta^{15}N_{PN}$ data should be verified with the nutrient data in future studies. Furthermore, the

POM investigated was weakly influenced by the terrestrial OM supplied by the Yangtze River during summer 2013 due to the reduced sediment supply by the Yangtze River and north-eastward transport of riverine particles to the northern East China Sea. We demonstrated that the composition of POM around DCM layers in the southern East China Sea is highly dynamic and largely driven by phytoplankton abundance. Nonetheless, additional ~~data of~~ radiocarbon and biomarker~~s~~ data are ~~crucial~~ needed to ~~revalidate~~re-evaluate whether or not the POM around the DCM water depths is influenced by terrestrial OM in the river-dominated East China Sea.

1 Introduction

Stable isotopes of organic carbon and nitrogen (δ^{13}C, δ^{15}N) and molar carbon to nitrogen (C/N) ratios are natural tracers~~the most~~ frequently used ~~natural tracers~~ to identify the source and fate of terrestrial organic matter (OM) in the estuarine and marine environments (Meyers, 1994; Hedges et al., 1997; Goñi et al., 2014; Selvaraj et al., 2015). This approach is based on the significant difference in δ^{13}C, δ^{15}N and C/N ratios between different end-members~~, especially~~ (e.g., terrestrial and marine), and the assumption that only a physical mixing of OM from compositionally distinct end-members occurs in these marginal settings (Thornton and McManus, 1994; Hedges et al., 1986). Quantifying ~~fractions~~ the relative contributions of end-members ~~by~~ using mass balance models thus requires known and constant ~~values of~~ elemental and isotopic values of end-members ~~of~~ and major sources of OM in the study region~~to the depositional system~~ (e.g., Goñi et al., 2003). ~~Any study applying~~ Therefore, application of mixing models for the discrimination of OM source~~s~~ ~~discrimination should therefore~~ requires clearly identified~~identify~~ representative values for ~~the~~ local OM sources~~of OM inputs into the area under investigation~~. However, in most cases, end-member values of δ^{13}C, δ^{15}N and molar C/N ratios ~~were simply replaced~~ are represented by 'typical' numbers, such as ca. –20 ‰ and –27 ‰ for δ^{13}C of marine phytoplankton and terrestrial plants (Kandasamy and Nagender Nath 2016 and references therein), respectively, but without measuring discrete end-member values in real, local or regional OM source materials. For example, ~~isotopic values of marine phytoplankton have not been measured in a number of earlier studies that employed~~ a number of earlier studies failed to measure isotopic values of marine phytoplankton despite using end-member mixing models to distinguish marine versus terrestrial OM ~~organic matter~~ in surface sediments (e.g., Kao et al., 2003; Wu et al., 2013), or these numbers simply represented by values of particulate organic matter (POM) in surface waters in the studied system (e.g., Zhang et al., 2007) or elsewhere from other ocean basins (e.g., Hale et al., 2012). It is known that stable isotopes (δ^{13}C, δ^{15}N) and molar C/N ratios of POM in estuarine and marine areas are representative of ~~these values in~~ primary production-derived OM when POM are mostly derived from phytoplankton biomass ~~and in that they are largely synthesized by phytoplankton~~ (Gearing et al., 1984). Since phytoplankton are ~~is~~ the main primary producer of marine OM, the elemental and isotopic compositions of phytoplankton should therefore be considered while studying the dynamics of POM in the marine water column.

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Chlorophyll *a* (Chl *a*) concentration in sea water is often used as an index of phytoplankton biomass ~~and phytoplankton carbon~~ (Cullen et al., 1982; Malone et al., 1983). The deep chlorophyll maximum (DCM) layer, which contributes significantly to the total biomass and primary production in the whole water column (Weston et

5 al., 2005; Hanson et al., 2007; Sullivan et al., 2010), is approximately equal to the subsurface biomass maximum layer (e.g., Sharples et al., 2001; Ryabov et al., 2010). The formation of maximum chlorophyll concentration at the DCM layer has been explained by several mechanisms: the differential zooplankton grazing with depths (Riley et al., 1949; Lorenzen, 1967), adaption of phytoplankton to light intensities or to increased concentration of nutrients (Nielsen and Hansen, 1959; Gieskes et al., 1978; Hickman et al., 2012), chlorophyll accumulation by

10 sinking detritus of phytoplankton (Gieskes et al., 1978; Karlson et al., 1996), ~~and~~ decomposition of chlorophyll by light (Nielsen and Hansen, 1959), and wind-driven nitrate supply and nitrate uptake in seasonally-stratified shelf seas (Hickman et al., 2012; Williams et al., 2013). The DCM layer is common in both coastal and open oceans, occurring at relatively shallow depths (1–50 m) in coastal seas, but in deeper depths (80–130 m) in open ocean (Cullen, 1982; Gong et al., 2015), and often variable in time and space (Karlson et al., 1996). For example, the

15 DCM layers were reported at depths of 30–50 m across the shelf in the southern East China Sea during summer from 1991 to 1995 (Gong et al., 2010). Hence, $\delta^{13}C$, $\delta^{15}N$ and molar C/N ratios of POM in the DCM layers of the continental shelf waters should reflect the $\delta^{13}C$, $\delta^{15}N$ and molar C/N ratios of phytoplankton (Savoye et al., 2003; 2012; Gao et al., 2014).

20 East China Sea is one of the largest marginal seas in the world, receiving huge quantities of freshwater (905.1 $km^3 \, yr^{-1}$; Dai et al., 2010) and organic ~~carbon~~ C (2.93 Tg C yr^{-1}, Tg = 10^{12} g; Qi et al., 2014) from the Yangtze River (Changjiang). Nutrient-rich freshwater input~~s~~ in turn stimulate~~s~~ the water column productivity ~~significantly~~ in coastal waters compared to the open ocean. ~~The annual~~ Annual primary production ~~over~~ for ~~the~~ entire shelf of the East China Sea is high relative to other ~~among the~~ marginal seas and was ~~has been~~ estimated to be 85 Tg C

25 yr^{-1} in 2008 (Tan et al., 2011). Several studies have been carried out on the physical, chemical and biological aspects of the East China Sea, including distributions of seasonal currents (e.g., Gong et al., 2010), chemical hydrography and nutrient~~s~~ distribution (Chen, 1996, 2008) and phytoplankton species composition in the water column (e.g., Zheng et al., 2015; Jiang et al., 2015). Likewise, $\delta^{13}C$, $\delta^{15}N$ and molar C/N ratios of POM have been determined for ~~constrained in~~ a limited number of transects across the East China Sea (e.g., Wu et al.,

30 2003; 2007a) as well as in a wide area of the western North Pacific marginal seas (Chen et al., 1996). Nonetheless, studies on elemental ratios and stable isotopic compositions of POM in DCM layers in the continental shelf of the East China Sea, especially along the indirect transport pathway of the Yangtze-derived terrestrial material to the Okinawa Trough (Chen et al., 2017), are poorly studied ~~almost unavailable~~. ~~In a recent study, Gao et al. (2014)~~ A recent study in the northern East China Sea investigated elemental and isotopic

compositions of POM in the surface, DCM and bottom layers on both seasonal and inter-annual timescales (Gao et al., 2014);~~in different seasons and years, but they focused on the northern part of the East China Sea with a scanty~~ however, there was minimal attention ~~has been paid on the~~given to biogeochemical processes associated with ~~involved in~~ the DCM~~ layers~~. Here, we investigate δ^{13}C, δ^{15}N and molar C/N ratios of suspended POM around the DCM layer in the continental margin of the East China Sea, in particular the area south of the Yangtze estuary, aiming (1) to comprehend the sources of POM in DCM layers and (2) to understand the factors controlling δ^{13}C and δ^{15}N dynamics in DCM layers of the southern East China Sea.

2 Study area

The East China Sea (ECS; Fig. 1) is the largest river-dominated marginal sea in the north-western Pacific region (Chen et al., 2017). The ~~continental shelf of the~~ECS shelf is wide (>500 km), but relatively shallow (<130 m) with an average water depth of 60 m (Gong et al., 2003; Liu et al., 2006)~~, but wide (>500 km)~~. The Yangtze River (Fig. 1), with a catchment area of more than 1.94 × 10^6 km^2 (Liu et al., 2007), is the main source of freshwater and sediment to the continental shelf. It is the fifth largest river in terms of water discharge (900 km^3 yr^{-1}) and the fourth largest river in terms of sediment discharge (470 Mt yr^{-1}) in the world (Milliman and Farnsworth, 2011).

In addition to the huge inputs of nutrients (dissolved inorganic nitrogen-DIN: 61.0±13.5 × 10^9 mol yr^{-1} for the interval of 1981–2006; Chai et al., 2009) and sediments from the Yangtze River, the ECS is characterized by a complex circulation pattern that is largely driven by the seasonally reversing East Asian monsoon winds (He et al., 2014; Chen et al., 2017). The surface circulation in the shelf is characterized by the south-north China Coastal Current (CCC) in the west, northward-moving Taiwan Warm Current (TWC) in the ~~middle~~central part and the ~~north~~north-eastward-flowing Kuroshio Current (KC) in the east (Fig. 1) (Liu et al., 2006). The Changjiang Diluted Water (CDW) is a mixture of ~~freshwater of~~Yangtze River freshwater and the ~~shelf water of~~ East China Sea shelf water~~,~~ and is characterized by a low salinity (<30, Umezawa et al., 2014). Owing to a huge amount of freshwater discharge from the Yangtze into the ECS, it is thought ~~has been believed~~that the CDW is the main ~~source~~component of CCC (Fig. 1). Because of the East Asian monsoon, where there is a strong northeast monsoon in winter and a weaker southwest monsoon in summer, the CDW flows southward along the coastline of mainland China as a narrow jet in winter (Chen, 2008; Han et al. 2013), whereas the same spreads mainly to the northeast in summer (Isobe et al., 2004). The Taiwan Warm Current (TWC) is a mixture of the warm water from the Taiwan Strait and ~~the~~intruding saline Kuroshio water; the latter is thought to be the most dominant source of heat and salt to the ECS (Su and Pan, 1987; Zhou et al., 2015). In addition, ~~there is an upwelling of~~ Kuroshio Subsurface Water (KSSW) is upwelled in the northeast off Taiwan Island due to an abrupt change in~~of~~ seafloor topography ~~in the outer shelf of~~ at the ECS outer shelf (dashed ellipse in Fig. 1) (Su et al., 1989; Sheu et

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al., 1999). The upwelled, oxygen-unsaturated KSSW is characterized by low temperature, ~~but~~ high salinity and high nutrients (Liu et al., 1988; Wong et al., 1991). The water exchange rate between the ~~East China Sea~~ECS water and Kuroshio water was estimated to be about 22,000 ± 9000 km^{-3} yr^{-1}, which is approximately 25 times the amount of Yangtze runoff into the ECS (Li et al., 1994; Sheu et al., 1999). Furthermore, Kuroshio water

5 accounts for up to~~made up~~ 90% of the shelf water in the ECS (Chen, 1996; Sheu et al., 1999).

The primary productivity in the ECS is limited by nitrogen in summer, but light in winter (Chen et al., 2001; Chen and Chen 2003). With the highest primary production during summer, annual primary production showed distinct spatial and temporal variations of 155 g C m^{-2} yr^{-1}, 144 g C m^{-2} yr^{-1} and 145 g C m^{-2} yr^{-1} in the north-western

10 ECS, south-eastern ECS and the entire ECS, respectively, in 1998 (Gong et al., 2003). The primary productivity has however decreased by 86% between 1998 and 2003 due to a large number of impoundments in the drainage basin of Yangtze River (Gong et al., 2006).

3 Materials and methods

15 ### 3.1 Sample collection

To investigate the biogeochemical characteristics of POM in the DCM layer of the southern East China Sea, suspended particles around the DCM water depths (10–130 m; Table 1) were collected from thirty-six stations along seven transects across the continental shelf by the *Science 3* cruise during summer (June 22–July 21)

20 2013 (Fig. 1). At each site, the physical properties of the water column were recorded by a Conductivity-Temperature-Depth (CTD) rosette (Seabird, SBE911+) fitted with a Seapoint chlorophyll fluorometer to detect the fluorescence maximum (see Supplementary Table S1 for the whole dataset). Sea water was collected using the rosette of Niskin water bottles attached with the CTD frame. and then stored in 5 L PVC bottles. All PVC bottles had been soaked in 0.1M HCl and then cleaned by distilled water. The volume of each water sample was

25 measured by graduated cylinder before filtration. Suspended particles were obtained by filtering 4.1–19.1 L of seawater collected around the fluorescence maximum layer through 0.7 µm/47 mm Whatman Glass Fiber Filters (GF/F), which were wrapped in aluminium foil. The filtration was under an ultimate pressure of 0.08 MPa to and avoided rupturing of phytoplankton cells (Steinman et al., 2017). All filters ~~have~~ had been pre-combusted at 450 ºC for 4 h in a muffle furnace to remove the background carbon and pre-weighed for determining the

30 concentration of suspended particulate matters (SPM). After filtration, filters were folded without rinsing and wrapped again in aluminium foil and then stored at –20 ºC immediately in a freezer onboard before they were brought back to the laboratory for further analysis.

3.2 Determination of SPM concentration and analyses of Chl *a*, POC, PN, $δ^{13}C$ and $δ^{15}N$

In the laboratory, filters with suspended particles were freeze-dried and then dried in an oven at 50 °C for 48 h. The weight difference between the dried filter and the same filter its counterpart before the filtration was used to calculate the weight of SPM. Five SPM samples (DH1-2, DH2-1, DH3-1, DH7-1 and DH7-7; Fig. S1) from water depths ranging between 20 m and 50 m were randomly selected for the measurement of chlorophyll a (Chl a) concentration. Chlorophyll a was extracted using 90% acetone and then determined spectrophotometrically according to Lorenzen (1967) and Aminot and Rey (2000). Briefly, the absorbance of sample extraction was measured at 665 nm and 750 nm against a 90% acetone blank before ($E665_o$, $E750_o$) and after ($E665_a$, $E750_a$) acidification with 1% HCl by the UV-Vis spectrophotometer (UV 1800, Shimadzu). Chl a concentration ($\mu g\ L^{-1}$) was calculated as: Chl a = 11.4 × 2.43 × (($E665_o$ − $E750_o$) − ($E665_a$ − $E750_a$)) × V_e /L × V_f, where V_e and V_f were the volumes of sample extraction and sea water filtered (ml), respectively, and L was the cuvette light-path (cm) (Aminot and Rey, 2000).

Prior to the measurement of POC and PN contents and their stable isotope values ($\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$) in SPM samples, a half of each filter was placed in a culture dish and 3 ml of 1N HCl was then added into the dish by a dropper and allowed them to react for 16 h to remove inorganic carbon (mainly carbonate). De-carbonated sample was dried at 50 °C for 48 h in an oven for HCl evaporation. Then a half of the de-carbonated filter (i.e. a quarter of the original filter, ~11 mm) was then punched and placed in tin capsules for further analysis. The POC and PN contents and their $\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$ compositions were measured at the Stable Isotope Facility of University of California Davis in USA, by using an elemental analyser (EA) (Elementar Analysensysteme GmbH, Hanau, Germany) interfaced to a continuous flow isotope ratio mass spectrometer (IRMS; PDZ Europa 20–20, Sercon Ltd., Cheshire, UK). During the isotopes ($\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$) analyses, different working standards (Bovine Liver, Glutamic Acid, Enriched Alanine and Nylon 6) of compositionally similar to the samples were used and were calibrated against NIST Standard Reference Materials (IAEA–N1, IAEA–N2, IAEA–N3, USGS–40, and USGS–41). The standard deviation is was 0.2 ‰ for $\delta^{13}C$ and 0.3 ‰ for $\delta^{15}N$. Isotopic values were presented in standard δ-notation as per mil deviations relative to the conventional standards, i.e. VPDB (Vienna Pee Dee Belemnite) for carbon and atmospheric N_2 for nitrogen, that is δX (‰) = [(R_{sample} − $R_{standard}$)/$R_{standard}$] × 10^3, where X = ^{13}C or ^{15}N, R = $^{13}C/^{12}C$ or $^{15}N/^{14}N$, R_{sample} and $R_{standard}$ are the heavy (^{13}C or ^{15}N) to light (^{12}C or ^{14}N) isotope ratios of sample and standard, respectively (e.g., Selvaraj et al., 2015).

Lorrain et al. (2003) cautioned that the measurement of PN and $\delta^{15}N$ after freezing increases the uncertainty of $\delta^{15}N$ and in combination with the concentrated HCl treatment, leads to a loss of PN and alteration of the $\delta^{15}N$ signature. Therefore, PN content and $\delta^{15}N$ values in the current study may have some bias due to de-carbonation. Nonetheless, similar methodological approach has been adopted by Wu et al. (2003) while

investigating suspended particles along the *PN* transect in the East China Sea (Fig. 1) and by Hung et al. (1996) while studying the suspended particles in the entire East China Sea. For instance, the range of δ^{15}N values (~3.8–8.4 ‰) obtained in the present study is comparable to the range of δ^{15}N values (ca. 0.7–9.4 ‰) obtained by Wu et al. (2003) for the entire water column. In addition, precision for δ^{13}C and δ^{15}N decreases for samples containing less than 100 µgC and 20 µgN, respectively. Among thirty-six filters analyzed for the present study, only five (three) filters contain less than 100 µgC (20 µgN).

4 Results and interpretations

4.1 Hydrographic characteristics and chlorophyll *a*

4.1.1 Temperature and salinity

Figure 2 illustrates the vertical distributions of temperature and salinity along seven transects across the ECS. ~~In the entire study area,~~ Water temperature in the upper 300-m ~~water column~~ varied from 15 ºC to 30 ºC, with ~~and~~ distinct ~~water column~~thermal stratification of the water column across the entire study area~~was evident from the temperature profiles~~ (Fig. 2). The temperature decreases when depth increases and the highest temperature (~30 ºC) seen mostly in the surface water and the lowest temperature (5 ºC) was observed in stations DH7–8 and DH7–9 at water depths of 850 m and 800 m, respectively (Fig. 2 and Table S1). Temperature at sampling depths of SPM ranged from 19.1 °C to 28.2 °C, showing a general decreasing trend from the inner to outer shelf in each transect (Fig. 2).

Salinity in general shows an increasing trend with water depths (Fig. 2), varying from 26.9 to 34.8 with an average value of 34.6 for the entire water column. An increasing trend of salinity from the west to east is evident in all seven transects (Fig. 2). The low salinity (<30) was constrained in the upper 10 m in four coastal stations (DH1–1, DH2–1, DH3–1, CON02; Fig. 2), wherein temperature is <24 ºC, indicating the limited influence of CDW plume in the study area. The middle salinity (30<S<34.1) was observed at a depth interval between 10 m and 30 m in stations (DH1–1, DH1–2, DH2–1, DH2–2, DH3–1; Fig. 2), but it spreads to a depth interval between surface and 30 m in the remaining stations. High salinity was mostly prevalent at bottom depths in all stations investigated. The salinity distribution at depths of SPM sampling shows an increasing trend from the inner to outer shelf (Fig. 2) and varied from 32.7 to 34.7 with an average salinity of 34.0, indicating low influence of CDW at DCM depths in the study area.

4.1.2 Turbidity

The turbidity in the water column of the ECS varied from 0.0 to 20.9 Formazin Turbidity Unit (FTU) (Fig. 3). In the inner shelf region, the vertical distribution of turbidity shows an obvious downward increasing trend and these high turbidity stations were limited along the coast (Fig. 3). This indicates sediment resuspension from the sea floor that was probably induced by hydrodynamic forces such as tides, waves and currents in the shallow coastal region. In the outer shelf stations, the turbidity was uniformly low from the surface to the bottom. Overall, most water depths where the SPM were sampled have low turbidity (<2.0 FTU), except for stations CON02 (4.75), DH5–1 (3.44), and DH7–1 (5.52) (Fig. 3).

4.1.3. Chlorophyll fluorescence and chlorophyll a (Chl a)

~~The concentration of Chl fluorescence varied up to18.0 µg L⁻¹ in the study area.~~ The highest Chl fluorescence concentration (18.0 µg L^{-1}) was observed in surface water~~s~~ at station DH3–1~~,~~ and all other values ~~are~~ were less than 8.0 µg L^{-1} (Fig. 3). The vertical profiles of Chl fluorescence usually show~~ed~~ a clear maximum in the subsurface layer at around 20 m in near coastal stations and 50 m in outer shelf stations (Fig. 3). The Chl fluorescence in the sampling depth ranged from 0.1 to 4.1 µg L^{-1}. Around 70 % of SPM sampled in this study falls in the DCM and/or contiguous to the DCM layer (open squares in Fig. 3), ideally representing the biogeochemical behaviours of POM ~~straddling around~~across the DCM layer. Based on the photosynthetically active radiation (PAR), we defined the euphotic depth~~,~~ as a depth at which the PAR is 1 % of its value at the sea surface and photosynthesis can take place (Kirk, 1994; Ravichandran et al., 2012; Guo et al., 2014a). The euphotic depth increased from the inner shelf (20 m) to the outer shelf (100 m) region. This is consistent with average euphotic depth of 33 m calculated based on the empirical relation: Z_{eu} = 4.605/K_d(PAR) (Kirk, 1994), where K_d(PAR) = 1.22K_d(490) (Tang et al., 2007; Ravichandran et al., 2012) and a mean value of 0.115 for K_d(490) for the East China Sea in summer was taken from Chen and Liu (2015). The presence of DCM layers near the euphotic depths suggests a close relationship between the light availability and deep chlorophyll maximum, and the OM in the SPM samples was likely to be dominated by the phytoplankton ~~productivity~~.

Linear correlation between the measured Chl a values and the fluorescence values obtained directly from the calibrated sensor attached with the CTD rosette is high with R^2 = 0.93 (see Fig. S1 in the Supplementary material). This relationship was used to convert the fluorescence values into Chl a concentration of all the remaining SPM using an equation: y = 0.708 x + 0.199, where y is Chl a concentration and x is *in situ* fluorescence value. The Chl a concentration varied from 0.28 to 3.08 µg L^{-1}. The highest value is observed in near coastal station DH5-1, whereas the lowest value is noted in station DH7-9 located off northeast Taiwan. The

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converted Chl *a* values were used to calculate the POC/Chl *a* ratio (Table S1), which is discussed in section 5.2.2.

4.2 POC and PN

The concentration of SPM ranged from 1.7 to 14.7 mg L^{-1} with a mean value of 4.4 mg L^{-1} (Table 1). The spatial distribution of SPM ~~shows~~ showed higher values in the inner shelf region and lower values in the outer shelf region (Fig. 4), consistent with the water column turbidity (Fig. 3). The POC concentration~~s~~ in the DCM layer varied between 20.4 and 263.0 µg L^{-1}, with a mean value of 85.5 µg L^{-1} (n = 36) (Fig. 4). The PN ranged from 4.4 to 52.8 µg L^{-1}, with a mean value of 17.7 µg L^{-1} (n = 36). The spatial distributions of POC and PN resemble each other (Fig. 4). The highest concentrations of POC (263 µg L^{-1}) and PN (52.8 µg L^{-1}) ~~are~~ were associated with station DH5-1 (Fig. 4 and Table S1). ~~Higher concentrations of~~ POC ~~(>90 µg L^{-1})~~ and PN ~~(>21 µg L^{-1})~~concentrations ~~are~~ were higher near the coast on the inner shelf (>90 µg L^{-1} and >21 µg L^{-1}, respectively)~~mostly observed in the inner shelf along the coastal line~~, ~~decreasing~~ and decreased gradually ~~towards the~~with distance offshore ~~direction~~ (Fig. 4). Lower concentrations of POC and PN are observed in the easternmost stations, ~~nearby off~~near northeast Taiwan Island (Fig. 4). Although the concentrations of both POC and PN varied by more than an order of magnitude (Fig. 4), the molar C/N ratios are fairly uniform at DCM layers ~~of the entire ECS~~throughout sampling, ranging from 4.1 to 6.3 with a mean ratio of 5.6±0.5 (n = 36) (Table 1).

4.3 δ^{13}C$_{POC}$ and δ^{15}N$_{PN}$

Spatial distributions of δ^{13}C$_{POC}$ and δ^{15}N$_{PN}$ around DCM layers are presented in Fig. 5. δ^{13}C$_{POC}$ decreased from the inner shelf to offshore region, varying widely from –25.8 ‰ to –18.2 ‰ (Table 1). Consistent with ~~to~~ the POC concentration, the highest δ^{13}C$_{POC}$ value (–18.2 ‰) is also associated with the coastal station DH5-1. The range of δ^{15}N$_{PN}$ is 4.2 ‰, varying between 3.8 ‰ and 8.0 ‰ (Table 1). The lowest δ^{13}C$_{POC}$ values (–25.8 ‰ and –25.2 ‰) ~~are found~~were observed northeast of Taiwan Island in the Okinawa Trough, ~~off northeast Taiwan Island, while the~~whereas δ^{15}N$_{PN}$ values (6.73 ‰ and 7.78 ‰) in ~~the same locations are~~this region were higher ~~(6.73 ‰ and 7.78 ‰)~~ than ~~that~~ those of the surrounding ~~location~~ area (Fig. 5). The spatial distribution of δ^{13}C$_{POC}$ ~~is~~ was quite similar to the spatial distribution of POC (Fig. 4), and the correlation coefficient (R^2) between δ^{13}C$_{POC}$ and POC was 0.55 (p<0.0001; Fig. 10).

5 Discussion

5.1 Influence of different water masses in the southern ECS

In order to identify the different water sources in the study area, temperature–salinity (*T–S*) diagrams were drawn for the entire water column (Fig. 6a) as well as for the SPM sampling depth around DCM layers (Fig. 6b). The *T–S* diagram for all the water depths shows a convergence at around 17 ºC, 34.6 (Fig. 6a), representing the upwelling of KSSW (Umezawa et al., 2014). There are two trends in the *T–S* diagram, indicating a mixing of three

5　water masses: one is less saline and much colder water, mainly CDW, another is more saline and warmer, mainly Taiwan Warm Current Water (TWCW), and the third one is KSSW (Fig. 6a). ~~The shelf water in the entire ECS in summer 2013 was mixed primarily by three water masses, CDW, KSSW, and TWCW (Fig. 6a).~~ The low salinity observed at five coastal sites (DH1-1, DH2-1, DH2-2, DH3-1 and CON02; Fig. 2) indicates the influence of CDW mostly in surface water, but also some of the DCM depths where water was sampled for SPM. This is

10　also evident from Fig. 6b where five stations fall within the area of shelf mixed water (SMW), which is a water body composed of a mixing between CDW and KSSW. However, except at these five coastal stations, most DCM depths where water was sampled for SPM seem to be weakly influenced by the CDW (Fig. 6b). Based on the *T–S* range of different water masses (Fig. 6), we further delineated the area ~~influenced along with~~and water depths influenced by three important water masses: CDW, TWCW and KSSW (Fig. 7). Interestingly, the influence

15　of CDW was constrained to ~~only in~~ the upper ~~0–~~10 m in five coastal stations ~~during the sampling time~~, whereas TWCW ~~influences around~~influenced the upper ~~0–~~30 m~~, covering~~ and covered three~~-fourths~~ quarters of the study region~~area~~, ~~and~~ with KSSW ~~seems to be~~largely ~~influenced~~influencing the bottom water across ~~of~~ the entire study region ~~area~~ (Figs. 2, 6a and 7).

20　In summary, although the river runoff was huge, the influence of CDW plume in the southern part of the ECS was weak during summer 2013 mainly because most of the CDW plume was transported ~~to northeastwardly~~northeastward of the Yangtze estuary to the Korean coast (Isobe et al., 2004; Bai et al., 2014; Gao et al., 2014). This contrasts with summer 2003 when the plume front moved southward (Bai et al., 2014). Meanwhile, the intrusion of TWCW and KSSW was strong in the continental shelf of the East China Sea during

25　summer 2013.

5.2 Characterization of POM in DCM layers

5.2.1 Molar C/N Ratio

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A necessary first step in the source analysis of POM using bulk carbon and nitrogen isotopes as well as the molar carbon to nitrogen ratio is to identify the form of total nitrogen in the measured SPM, so that inorganic nitrogen is not miss-assigned into nitrogenous organic endmember (Hedges et al., 1986). The linear relationship between POC and PN ($R^2 = 0.98$, $p<0.0001$; Fig. 8a) suggests that nitrogen is strongly associated with organic

carbon. The slope of linear regression of POC against PN corresponds to a molar C/N ratio of 5.76 (Fig. 8a). The positive intercept on the PN axis when POC is zero represent the amount of inorganic nitrogen (~0.03 µM), indicating that essentially all nitrogen are in the organic form. The molar C/N ratios of all SPM samples (4.1–6.3) from the DCM layers are lower than the canonical Redfield ratio (6.63) (Fig. 8a), but are similar to the average

5 molar C/N ratios of 5.6 for marine POM (Copin-Montegut and Copin-Montegut, 1983) and 6 for POM in cold, nutrient-rich waters at high latitudes (Martiny et al., 2013). The range also falls within the range of 3.8 to 17 reported for marine POM (Geider and La Roche, 2002), but it is higher than an unprecedented low C/N ratio (2.65±0.19) of POM in Canada Basin that was attributed to a dominant contribution of smaller size (<8 µm) phytoplankton to POC (Crawford et al., 2015). Wu et al. (2003) investigated the C/N ratio of POM (4.3–29.2) at all

10 depths along the *PN* transect, a standard cross-shelf section extending from the Yangtze estuary southeast to the Ryukyu Islands, crosscutting the Okinawa Trough and perpendicular to the principle axis of Kuroshio Current in the ECS (Fig. 1). Liu et al. (1998) measured the C/N ratio of POM in the surface water of the ECS and found a wider C/N ratio from 4.0 to 26.9 with a mean ratio of 7.6 in spring and from 4.7 to 34.3 with a mean ratio of 15.2 in autumn 1994. The authors attributed the lower C/N in spring to ~~an~~ more intense biological activity than in autumn,

15 and the spatial distribution of C/N was thought to be related to that of phytoplankton abundance.

Characteristically, a narrow range of low C/N ratios in our SPM samples and less influence of CDW in the study region (Fig. 7) confirm~~s~~ the lack of terrestrial signals transported mainly by the Yangtze River. We therefore suggest that the POM in the DCM layers of southern East China Sea is dominated by marine-sourced OM with

20 an unrecognized contribution of terrestrial OM. Low C/N ratios further restrict the assumption of degradation of nitrogen-rich OM, a process that normally increases the C/N ratio to more than that of the Redfield ratio. Therefore, the molar C/N ratio can be better explained as a source signal of OM rather than OM degradation in the SPM investigated in this study.

25 **5.2.2 POC/Chl *a* Ratio**

The linear correlation between POC and Chl *a* (R^2 = 0.49, p<0.0001; Fig. 8b) further indicates that the phytoplankton productivity is largely responsible for the POC production in the SPM samples. Moreover, the POC/Chl *a* ratio of 34.1 g g^{-1} derived from the slope of a regression line (y = 34.1 (±9.99) x +49.9 (±8.86) (Fig.

30 8b) is consistent with the reported POC/Chl *a* ratios in the ECS (36.1 g g^{-1}; Chang et al., 2003) and the North-western Pacific (48 g g^{-1}; Furuya, 1990). However, the POC/Chl *a* ratio obtained in this study is lower than that estimated (64 g g^{-1}) for the sinking particles in the ECS and the Kuroshio region, off northeast Taiwan Island (Hung et al., 2013). The range is well within the range (13–93 g g^{-1}) reported for POM in the ECS by Chang et al. (2003) and is also consistent with the range (18–94 g g^{-1}) estimated from phytoplankton cell volumes by the

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same authors. Although the Chl *a* concentration in our study was converted based on the linear relationship between measured Chl *a* and *in situ* fluorescence values (see Section 3.2 and Fig. S1 for more details), it is more or less similar to Chl *a* concentrations obtained in the above-mentioned studies, which were mostly extracted from filtered particles (Chang et al., 2003; Hung et al., 2013).

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POC/Chl *a* ratio has been used for the discrimination of POM sources in coastal ocean waters (Cifuentes et al., 1988). POC/Chl *a* ratio in living phytoplankton varies with temperature, growth rate, day length, phytoplankton species, and irradiance (Savoye et al., 2003 and references therein). The POC/Chl *a* ratio of living phytoplankton was reported to be between 40 and 140 g g^{-1} (Geider, 1987; Thompson et al. 1992; Montagnes et al. 1994; Head

10 et al. 1996). Furthermore, a POC/Chl *a* ratio of less than 200 g g^{-1} is an indication of a predominance of newly-produced phytoplankton (or autotrophic-dominated) in POM, and that a value higher than 200 g g^{-1} is an indication of detrital or degraded organic matter (or heterotrophic/mixture-dominated) (Cifuentes et al., 1988; Savoye et al., 2003; Liénart et al., 2016, 2017). The POC/Chl *a* ratio in the DCM layer of the ECS is almost <200 g g^{-1} (33–200 g g^{-1}), with one exception (CON02: 303 g g^{-1}; Fig. 9), indicating that POM in the DCM layers of

15 ECS was dominated by phytoplankton, as also indicated by the low C/N ratios (4.1–6.3). The relatively high POC/Chl *a* ratio only in one station, CON02 (Fig. 9), suggest that the POM in this sample was likely sourced from degraded phytoplankton OM, terrestrial OM, or heterotrophic-dominated OM. However, the molar C/N ratio of CON02 (5.3) is lower than the canonical Redfield ratio (6.63), eliminating the probability of degraded and terrestrial OM sources. In addition, the insignificant linear correlation between C/N ratio and POC/Chl *a* ratio (Fig.

20 9) supports the non-degraded POM, a process resulting in a simultaneous increase of C/N and POC/Chl *a* ratios, mainly because of the preferential decomposition of N-rich OM, as well as a fast degradation of Chl *a* than the bulk POC pool (e.g., Savoye et al., 2003). Thus, the POM in CON02 seems to be dominated by heterotrophic biota, though the exact reason for the dominance of heterotrophic biota only at one location in our study area is unknown and needs further investigation.

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Briefly, several clues indicate the predominance of newly-produced, phytoplankton-synthesized OM around DCM layers of the southern East China Sea: 1) low influence of fresh water, 2) low molar C/N ratios, 3) a linear correlation between POC and chlorophyll *a*, and 4) low POC/Chl *a* ratios, mostly <200 g g^{-1}.

30 **5.3 Dynamics of δ^{13}C$_{POC}$ in POM in DCM**

Although a narrow range of molar C/N ratio in the SPM indicated an aquatic origin for the POM at DCM layers, the wide variability of δ^{13}C$_{POC}$ (–25.8 to –18.2 ‰) suggests that the POM around DCM layers would be a mixture of terrestrial C3 plants with a typical δ^{13}C value of ca. –27 ‰ (e.g., Peters et al., 1978; Wada et al., 1987) and

marine phytoplankton with a typical $\delta^{13}C$ range of –18 to –20 ‰ (e.g., Goericke and Fry, 1994). However, Fig. 5 illustrates a distinct decreasing trend of $\delta^{13}C_{POC}$ towards the outer shelf; a pattern opposite to an increasing trend of $\delta^{13}C$ evident in suspended particles and surface sediments, i.e. seaward decrease of terrestrial OC in surface sediments of many river-dominated margins (Emerson and Hedges, 1988; Meyers, 1994; Hedges et al., 1997;

5 Kao et al., 2003; Wu et al., 2003). Such a spatial distribution with less negative $\delta^{13}C_{POC}$ values in the coastal region, but more negative $\delta^{13}C_{POC}$ values in the middle-outer shelf is inconsistent with the idea of terrestrial OC influence. The elevated $\delta^{13}C_{POC}$ values (average of –20.7 ‰) in the coastal region, concomitant with high POC concentrations (Fig. 4), are consistent with the higher marine primary productivity (11 g C m^{-2} yr^{-1}) reported in the western than that in the eastern parts of East China Sea (Gong et al., 2003). The lower $\delta^{13}C_{POC}$ occurred in the

10 middle-outer shelf region where oligotrophic Taiwan Warm Current Water and Kuroshio Water spread (Fig. 5). The lowest $\delta^{13}C_{POC}$ (–25.8 ‰) was observed at a water depth of 85 m, off northeast Taiwan, likely due to the intrusion of Kuroshio Subsurface Water with low $\delta^{13}C$ from –31 ‰ to –27 ‰ (Wu et al., 2003), is also in agreement with the hydrographic parameters of this location (Figs. 2 and 7).

15 A positive linear correlation between $\delta^{13}C_{POC}$ and POC ($R^2 = 0.55$, p<0.0001; Fig. 10a), a characteristic feature of productive oceanic regions (Savoye et al., 2003), suggesting the effect of growing primary productivity (and or increasing cell growth rate) on a decrease of carbon fractionation during photosynthesis (Miller et al., 2013). This is likely because of a limitation of dissolved CO_2, which cannot be compensated in time by the surrounding water in a relatively closed system because of stratification (Kopczyńska et al., 1995). Further, high productivity makes

20 ^{13}C-enriched OM in phytoplankton (Fry and Wainwright, 1991; Nakatsuka et al., 1992; Miller et al., 2013). Lowe et al. (2014) observed increased $\delta^{13}C$ and fatty acid concentration in the POM while increasing phytoplankton abundance in the nearshore waters of San Juan Archipelago, WA. Although primary productivity has a significant correlation with $\delta^{13}C_{POC}$, only 55 % of $\delta^{13}C_{POC}$ variation can be explained by primary productivity (Fig. 10a), implying that other factors, such as species and sizes of phytoplankton, must have influenced $\delta^{13}C$ values of

25 phytoplankton living in the DCM layers (Falkowski, 1991; Hinga et al., 1994).

The distribution of phytoplankton community in the East China Sea is affected by physicochemical properties (temperature, salinity and nutrients) of different water masses and surface currents (Umezawa et al., 2014; Jiang et al., 2015). Diatoms and dinoflagellates are the main phytoplankton communities in summer with 136 taxa of

30 diatoms from 55 genera and 67 taxa of dinoflagellates from 11 genera have been reported, along with minor communities of chrysophyta, chlorophyta and cyanophyta (Guo et al., 2014b). There is a clear decreasing trend of phytoplankton abundances in the East China Sea from the surface to bottom, as well as from the coastal to offshore region that is widely believed to be due to nutrient availability (Zheng et al., 2015). The phytoplankton species have distinct spatial characteristics, but no significant differences in species between surface waters and

the DCM layers (Zheng et al., 2015). Diatoms with large cell sizes were the dominant species in the coastal region, while phytoplankton with small sizes was dominant in the oligotrophic offshore shelf and Kuroshio waters (Furuya et al., 2003; Zhou et al., 2012). According to Jiang et al (2015), the contribution of micro- (>20 μm), nano- (3–20 μm) and pico-phytoplankton (<3 μm) to Chl a, respectively, was 40 %, 46 % and 14 % in nutrient-rich inshore waters, and 14 %, 34 %, and 52 % in offshore regions in summer 2009. The outer shelf region was composed of small size phytoplankton, mainly cyanobacteria and cryptophytes transported by Taiwan Warm Current and Kuroshio Current. It has been reported that diatoms have higher $\delta^{13}C$ values (–19 to –15 ‰) than dinoflagellates (–22 to –20 ‰; Fry and Wainwright, 1991; Lowe et al., 2014). Likewise, large phytoplankton have higher $\delta^{13}C$ values than small phytoplankton and heterotrophic dinoflagellates have higher $\delta^{13}C$ values than autotrophic dinoflagellates (Kopczyńska et al., 1995). Similarly, wide variations of $\delta^{13}C_{POC}$ (–22.05 to –27.62 ‰) at DCM layers in the northern East China Sea were documented by Gao et al. (2014). Significant variations of $\delta^{13}C$ in suspended OM that was dominated by phytoplankton were reported from the Delaware estuary (–25 to –20 ‰; Cifuentes et al., 1988), the Bay of Seine (–24.3 to –19.7 ‰; Savoye et al., 2003), the Santa Barbara Channel (Miller et al., 2013) and the nearshore waters of San Juan Archipelago, WA (–24.1 to –18.9 ‰; Lowe et al., 2014). These variations were influenced largely by the isotopic fractionation during phytoplankton photosynthesis and degradation than by changes in the relative contributions of terrestrial and aquatic OM (Fogel and Cifuentes, 1993; Savoye et al., 2003).

5.4 Temperature effect on the $\delta^{13}C_{POC}$ around the DCM layer

Apart from primary production and the growth rate and species composition, temperature and biomass degradation may influence the carbon isotopic composition of phytoplankton (Savoye et al., 2003). Temperature has an indirect effect on isotopic fractionation between phytoplankton carbon and dissolved CO_2, and therefore on phytoplankton $\delta^{13}C$ (e.g., Rau et al., 1992; Savoye et al., 2003). The C/N ratio, POC/Chl a ratio and $\delta^{13}C_{POC}$ all indicated that the POM around the DCM layer is dominated by newly-produced phytoplankton OM (see Sections 5.1–5.3). Therefore, to understand the temperature effect on $\delta^{13}C$ of phytoplankton, we plotted our $\delta^{13}C_{POC}$ data against temperature into two groups by separating approximately at ~24°C (Fig. 11a). Data points of both groups show a decreasing $\delta^{13}C$ of phytoplankton biomass ~~while~~ with increasing temperature around the water depths of DCM in the southern ECS (Fig. 11a). Such a relationship is in contrast to the positive relationship between these two variables observed for the surface ocean POM around the world (Sackett et al., 1965; Fontugne, 1983; Fontugne and Duplessy, 1981).

The negative relationship between $\delta^{13}C_{POC}$ and temperature is likely related to biological activity and carbonate dissolution equilibrium, both may control the concentration of dissolved inorganic carbon in the DCM layers,

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which are closer to euphotic depths (see Section 4.1). The weak correlation between $\delta^{13}C_{POC}$ and temperature supports a weak influence of temperature on $\delta^{13}C_{POC}$ around DCM layers in the study area (Fig. 11a). A decrease in fractionation of approximately −0.56‰ °C$^{−1}$ is estimated for POM collected at <24°C, whereas a decrease in fractionation of roughly −0.51 °C$^{−1}$ is estimated for POM collected at >24°C (Fig. 11a). In order to distinguish the influence of biological parameters from temperature on $\delta^{13}C_{POC}$, the $\delta^{13}C_{POC}$ data were corrected for the 'temperature effect' by normalizing the data using an equation: $\delta^{13}C_{POC} = f(T)$.

In the present study, since most $\delta^{13}C_{POC}$ values come from the DCM layer and the $\delta^{13}C_{POC}$ is negatively correlated with temperature (Fig. 11a), we applied our own temperature coefficients (−0.56‰ °C$^{−1}$ and −0.51‰ °C$^{−1}$) and $\delta^{13}C_{POC}$ was normalized at 24°C (i.e. the mean temperature at sampled water depths) using the formula (Savoye et al., 2003): $\delta^{13}C_{24°C} = \delta^{13}C_{POC} - s(T - 24)$, where $\delta^{13}C_{24°C}$ is the temperature-normalized $\delta^{13}C_{POC}$, T is the seawater temperature in °C from water depths where SPM sampled, and s is the slope of the linear regression $\delta^{13}C_{POC} = f(T)$ in ‰ °C$^{−1}$ obtained from Fig. 11a. There are significant correlations between $\delta^{13}C_{24°C}$ of biomass and POC concentration (circles: $R^2 = 0.71$; p<0.0001; n = 18 and triangles: $R^2 = 0.66$; p<0.0001; n = 18; Fig. 11b), indicating that primary production drives ~70% of the variation of phytoplankton $\delta^{13}C$ around DCM layers in the southern ECS. Similar positive relationship between temperature-normalized $\delta^{13}C$ and POC concentration was observed by Savoye et al. (2003) during spring phytoplankton blooms in the Bay of Seine, France. On the other hand, $\delta^{13}C_{24°C}$ correlated insignificantly with POC/Chl a ratio and C/N ratio (Figs. 11c and 11d), implying that degradation has a minor effect on the carbon isotopic composition of POM in this study.

5.5 Dynamics of $\delta^{15}N_{PN}$ in POM in DCM layers

In contrast to the POC and $\delta^{13}C_{POC}$ relationship (Fig. 10a), there is no significant relationship between PN and its isotopic composition ($\delta^{15}N_{PN}$) of the POM investigated in the present study (Fig. 10b), implying that primary productivity has no significant control on the variability of $\delta^{15}N_{PN}$. As the POM around the water depths of DCM was dominantly from the newly-produced, phytoplankton-synthesized source, $\delta^{15}N_{PN}$ should be similar to $\delta^{15}N$ in phytoplankton. Considering the prevalence of low N/P ratio in the DCM layer of the East China Sea (Lee et al., 2016), the degree of nitrate utilization by phytoplankton should be high and that would result in the composition of $\delta^{15}N_{PN}$ similar to $\delta^{15}N$ of nitrate ($\delta^{15}N_{NO3}{}^-$) (Altabet and Francois, 1994; Minagawa et al., 2001). Therefore, the spatial distribution of $\delta^{15}N_{NO3}{}^-$ is probably crucial to decipher the distribution of $\delta^{15}N_{PN}$ in DCM layers. Importantly, the spatial distribution of $\delta^{15}N_{PN}$ (Fig. 5) resembles the surface current pattern (Fig. 1), as well as the distribution of different water masses (Fig. 7), suggesting that nitrate and the $\delta^{15}N_{NO3}{}^-$ of CDW, TWCW and Kuroshio Water are largely governing the distribution of $\delta^{15}N_{PN}$ in the study area.

According to Li et al. (2010), the range of $\delta^{15}N_{NO_3^-}$ in the Yangtze River was 7.3–12.9 ‰, with a mean value of 8.3 ‰. In the northeast of Taiwan Island, $\delta^{15}N_{NO_3^-}$ was 5.5–6.1 ‰ at depths of 500 m to 780 m (Liu et al., 1996). However, TWCW is nutrient-depleted, enabling incorporation of N-fixer derived nitrogen in the suspended POM. This general spatial pattern of $\delta^{15}N_{NO_3^-}$, i.e. higher $\delta^{15}N_{NO_3^-}$ (>6 ‰) in the northeast coastal region and off
5   northeast Taiwan, but lower $\delta^{15}N_{PN}$ in between these two regions, exactly resembles the distribution of $\delta^{15}N_{PN}$ in the DCM layers of this study (Fig. 5). Therefore, the $\delta^{15}N_{PN}$ variation in the DCM layer of the East China Sea was primarily governed by the nutrient status and $\delta^{15}N_{NO_3^-}$, though we do not have nutrient data generated during the same cruise to validate our interpretations.

10   There is another possibility that high $\delta^{15}N_{PN}$ (DH7-8: 6.7 ‰, DH7-9: 7.8 ‰) in the DCM layer, off northeast Taiwan (Fig. 5), may not ~~be resulted~~result from the high degree of nitrate utilization, but instead from the incorporation of inorganic nitrogen (mainly NH_4^+) in the POM. According to Chen et al. (1996) and Liu et al. (1996), NO_3^- and NH_4^+ concentrations in KSSW were high due to the decomposition of OM in sinking particles. However, the concentrations of Chl fluorescence as well as POC and PN are low (Figs. 3 and 4). The low Chl fluorescence
15   might be limited by the low temperature in this high nutrient low chlorophyll region (Umezawa et al., 2014). Because of the low temperature, the prevailing high CO_2 pressure expected to decrease $\delta^{13}C$ in DIC ~~that~~and may drive a great carbon isotopic fractionation during carbon assimilation by phytoplankton (Rau et al., 1992), the potential reason why $\delta^{13}C_{POC}$ values in these two stations ~~are~~were low (−25.8 ‰ and −25.2 ‰) compared to values of other locations in the study area. Consistently, the low concentration of POC restricts the idea that the
20   high $\delta^{15}N_{PN}$ could not be from the denitrification effect. The high $\delta^{15}N_{PN}$ (6.7 ‰, 7.8 ‰) are probably due to the incorporation of inorganic nitrogen (mainly NH_4^+), the process normally drives the $\delta^{15}N_{PN}$ as high as that of inorganic nitrogen $\delta^{15}N$ (Coffin and Cifuentes, 1999). Although $\delta^{15}N$ of NH_4^+ in Kuroshio Water is not available for comparison, it seems that $\delta^{15}N$ of remineralized NH_4^+ was relatively greater than $\delta^{15}N$ of NO_3^- (York et al., 2010). This possibility is also supported by the high concentrations of NO_3^- and NH_4^+ in Kuroshio Subsurface Water (Liu
25   et al., 1996) as well as the low contents of POC (<1 %; 0.96 %, 0.98 %) and low molar C/N ratios (4.1, 5.4) of these two SPM samples (DH7-8 and DH7-9).

5.5 Impact of Yangtze River on POM in DCM of ECS

30   The range of POC/Chl *a* obtained in this study (33–200 g g^{-1}) is within the range (<200 g g^{-1}) reported for the phytoplankton-dominated POM in the coastal and shelf waters (e.g., Chang et al., 2003; Savoye et al., 2003; Hung et al., 2013; Liénart et al., 2016). We also obtained a narrow range of C/N ratio (4.1–6.3), but a wide range of $\delta^{13}C_{POC}$ (−25.8 to −18.2 ‰) compared to previous studies in the ECS (4.0–34.3, Liu et al., 1998; −24.0 to −19.8 ‰~~ ‰~~, Wu et al., 2003). ~~These~~ Our results indicate~~d~~ that POM ~~around the water depth of~~at the DCM was largely

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~~derived from the synthesis of~~produced *in situ* ~~phytoplankton~~ and derived from phytoplankton biomass, ~~the influence of~~ with little terrestrial influence~~OM supplied by the Yangtze River to the ECS is low~~. The lack ~~missing~~ of terrestrial OM signals seems to be related to reservoir and dam buildings along the river in recent years that has shifted the location of the Yangtze-derived POC deposition from the inner shelf of the ECS to terrestrial reservoirs (Li et al., 2015). The sediment delivered from the river to the estuary has been reduced by 40 % since 2003 when the Three Gorges Dam (TGD) was completed (Yang et al., 2011 and references therein). Recently, Dai et al. (2014) reported that the ~~particles~~ particulate load discharged by the Yangtze has declined to 150 Mt yr^{-1}, less than ~70% of its sediment delivery to the ECS during 1950s. Although 87 % of the mean annual sediment of Yangtze River is discharged during the flood season from June to September (Wang et al., 2007; Zhu et al., 2011), approximately 60 out of 87% of the fine-grained sediments are temporarily deposited near the estuary and then later resuspended and transported southward along the inner shelf, off the mainland China (Chen et al., 2017 and references therein). The Yangtze-transported POM moves up toward the northeast across the shelf along the so called the Changjiang transport pathway in summer season (e.g., Gao et al., 2014), which is largely affected by the combined effects of high river discharge, southwest summer monsoon and the intensified TWC (Beardsley et al., 1985; Ichikawa and Beardsley, 2002; Lee and Chao, 2003). The *T–S* diagrams (Figs. 6 and 7) of this study also illustrate this view.

Accompanying ~~with~~ the decreasing sediment input, dam building in the Yangtze River basin since 2003 has buried around 4.9±1.9 Mt yr^{-1} biospheric POC, approximately 10% of the world riverine POC burial flux to the oceans (Li et al., 2015). The POC flux from the Yangtze to the ECS (range: 1.27–8.5 × 10^{12} g C yr^{-1}; Wang et al., 1989; Qi et al., 2014) was significantly less than the estimated primary productivity (72.5 × 10^{12} g C yr^{-1}; Gong et al., 2003), implying the predominance of marine-sourced organic matter in the ECS. Moreover, the substantial quantity of organic substances that transported by the Yangtze River may be completely modified before being ultimately deposited on the inner shelf of the ECS and being transported further offshore (Katoh et al., 2000; Lie et al., 2003; Chen et al., 2008; Isobe and Matsuno, 2008). Wu et al. (2007b), for instance, observed an advanced stage of POM degradation in the entire Yangtze River with an average degradation index of –1.1. Based on the investigation of lipid biomarkers in a sediment core collected from the ECS, Wang et al. (2016) suggested the dominant preservation of marine autochthonous organic matter (~90 %) in the ECS.

Summary and conclusions

In this study, we comprehensively characterized the particulate organic matter (POM) collected from the deep chlorophyll maximum (DCM) layer in the southern East China Sea using hydrographic data (temperature, salinity and turbidity), fluorescence (chlorophyll *a*) as well as elemental (POC, PN) concentrations and isotopic

($\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$) compositions. All these parameters indicated that the POM around DCM layers was dominantly composed of newly-produced OM by phytoplankton with a weak contribution from terrestrial input despite the study area is being the best example for the river-dominated continental margin in the world. We also discussed the main factors controlling the $\delta^{13}C$ and $\delta^{15}N$ variations in phytoplankton in the study area. As for the

5    $\delta^{13}C_{POC}$, the variations in primary productivity, as indicated by the positive correlation between $\delta^{13}C_{POC}$ and POC, and phytoplankton species were the main factors; the former explained ~70% of the variability in $\delta^{13}C_{POC}$, after ~~accounted~~ accounting for temperature effects. On the other hand, $\delta^{15}N_{PN}$ variation seems to be related to uptake of nitrate or locally regenerated ammonia, ~~but~~ which needs to be substantiated by the nutrient data in future studies. Our results show that phytoplankton dynamics drive marine POM composition around DCM layers in the

10   southern East China Sea.

Moreover, phytoplankton in the southern East China Sea contain relatively lower $\delta^{13}C_{POC}$ values than that of typical marine phytoplankton (–18 to –20 ‰). This emphasizes the need of sufficient investigation of end-member variability, which is crucial for the estimation of relative contributions of terrestrial and marine OM by end-member

15   mixing model. Therefore, our results with highly variable $\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$ values in the autotrophic-dominated DCM layers can provide unique ranges for these two isotopes in the East China Sea, especially the region south of 29 °N, and form a basis for the long-term evaluation of organic carbon burial along the inner shelf mud-belt, which is largely accumulated in the East China Sea during the Holocene.

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Table 1. Summary statistics of elemental and isotopic compositions, as well as C/N and POC/Chl *a* ratios, of suspended particulate matters (SPM) around DCM layers in the southern East China Sea (n=36). Chl *a* is the converted value using the linear relationship between measured Chl *a* and Chl Fluorescence. SD=Standard deviation.

	Sampling Depth (m)	SPM (mg L^{-1})	POC (µg L^{-1})	PN (µg L^{-1})	$\delta^{13}C_{POC}$ (‰)	$\delta^{15}N_{PN}$ (‰)	C/N Molar	POC/Chl *a* (g g^{-1})
Min	10	1.7	20.4	4.4	−25.8	3.8	4.1	33.3
Max	130	14.7	263.0	52.8	−18.2	8.0	6.3	303.3
Mean	45	4.4	85.5	17.7	−23.0	6.1	5.6	100.3
SD	21	2.7	49.5	9.9	1.5	1.0	0.5	51.8