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

Referee 1 (Prof. P.A. Meyers)

Referee 1: Liu and colleagues present 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. They measured carbon and nitrogen concentrations and isotopic compositions of 36 samples collected from 7 cross-shelf transects and augmented these data with a suite of standard hydrographic measurements. These data allowed them to conclude that little land-derived organic matter contributes to the suspended particulate matter despite the proximity of the sampling locations to the mouth of the Yangtze River. Instead, the organic matter freshly produced by phytoplankton. The authors attribute variations that they found in the carbon and nitrogen isotopic values of the organic matter to local differences in productivity rates and differences in the nitrate isotopic signatures of the major water masses in the area. The study seems to have been designed well, and the authors seem to have interpreted their results properly, but problems with the presentation make this contribution hard to read and appreciate. The English badly needs refining, and some additional details should be addressed.

Reply: Thank you very much for your appreciation on the overall performance of the research work that presented in the manuscript. We took the utmost care to refine our English in the revised version.

Referee 1: For a start, the second paragraph of the Introduction seems to be missing something (lines 27-32).

Reply: It seems that there is no link between the last two sentences of the first paragraph and the first sentence of the second paragraph and therefore this part has been revised as follows (**P3, L3-17 in the marked-up manuscript**):

It is known that stable isotopes (δ^{13} C, δ^{15} N) and molar C/N ratios of POM in estuarine and marine environments are representative of these values in primary production-derived OM and in that they are largely synthesized by phytoplankton (Gearing et al., 1984). Since phytoplankton 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.

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 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).

Referee 1: After that, Section 3.1 on sample collection should include a tabulation of the 36 samples that shows their water depths and some of their hydrographic properties. This tabulation could be an appendix, but the information that it would contain should be available to interested readers.

Reply: Table S1 is included in the appendix in the revised version. Table S1 includes station, coordinates, depth, hydrographic parameters (temperature, salinity and turbidity), chlorophyll fluorescence concentration and elemental (POC, PN) and isotopic (δ^{13} C and δ^{15} N) compositions of POM at water depths where the SPM sampled for the present investigation.

Referee 1: Then, the explanation for higher δ^{13} C and C/N values in surface sediments that phytoplankton (page 14, lines 10-16) seems out of place. This contribution is about POM, not sediments.

Reply: We agree with the comment from Referees 1 and 2, and thus this part has been deleted in the revision.

Referee 1: To continue with details that need correction, neither Table 1 nor Table 2 contribute much to the paper as they exist. I suggest either expanding Table 1 as suggested above or deleting it and providing a detailed appendix. The figures are effective, but Figure 3 could be improved by inverting the salinity color code so that salinity (and hence density) increases downward and Figure 8 needs to have the spelling of Redfield corrected the left panel and in the legend.

Reply: To understand the range of parameters studied, Table 1 has been retained in the revised version. However, as suggested by the Referee, Table S1 with the hydrographic, elemental and isotopic data is also included in the appendix.

As suggested, Table 2 has been deleted in the revised revision. Figure 3 has been improved; however, we don't know how to invert the salinity colour code using Ocean Data View. The spelling of Redfield has been corrected in Figure 8.

Thank you very much.

Anonymous Referee #2

General overview

The manuscript of Liu et al. is focused on characteristics of suspended particulate organic matter (SPOM) in the deep chlorophyll maximum (DCM) of the Eastern China Sea (ECS) during summer 2013. It is based on bulk descriptors of SPOM (C/N and POC/Chl a ratios as well as δ^{13} C and δ^{15} N). The mains findings are: 1) DCM SPOM mainly originates from in situ primary production, 2) terrestrial POM slightly or insignificantly contributes to DCM SPOM composition, and 3) the latter is contradictory to previous studies but illustrates the drastic decrease in the contribution of the terrestrial POM originating from the Yangtze River to the SPOM composition in the ECS. These findings are sounded and clearly illustrated by the present data set.

The manuscript is well organized and usually well illustrated.

It is of broad audience for scientist who are interested in organic matter cycling and land-to-sea export. It is within the scope of BG. However there are some issues in the present version of the manuscript that preclude the acceptance of the manuscript in its present version. These issues are:

- a lack of information in the methods

- many unneeded details and miscellaneous information that are not needed in the discussion, that rend the discussion too wordy and that dilute the main messages of the study. Authors should focus on what the data indicate, which is usually very clear.

- interpretations of $\delta^{15}N$ data set that are not correct or at least very partial. This data set cannot be published within this manuscript without deeply reconsidering its interpretation and without additional data set regarding N-nutrient (at least nitrate and ammonium) concentrations.

There are also some inconsistencies and language errors that have to be corrected. Thus, I recommend major revision

Reply: Thank you very much for your appreciation on the overall performance of the manuscript and critics on the discussion part and interpretations of $\delta^{15}N$ data.

Detailed comments

Referee 2: Section 3: lack of information and details needed

Conversion of fluorescence into chlorophyll a concentration: Since Chl a is a key parameter of the study and is used to calculate POC/Chl a ratio (which values are compared to reference values), it should be explained how in situ fluorescence was converted into Chl a concentration.

Reply (**P6**, **L15-18**): 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).

(**P9, L7-13**) 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 $R^2 = 0.93$ (Fig. S1 in the Supplementary material). This relationship was used to convert the fluorescence values to Chl *a* concentrations of all the remaining SPM using an equation: $y = 0.708 \times + 0.199$, where y is Chl *a* and x is chlorophyll fluorescence concentration. 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 converted Chl *a* values were used to calculate POC/Chl *a* ratio (Table S1), which is discussed in section 5.2.2.

Referee 2: Section 3.1, line 9: indicate the range of depths of the samplings for SPM.

Reply (P5, L27-30): The sentence has been revised as follows:

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 Cruise during summer (June 22–July 21) 2013 (Fig. 1).

Depth of each station sampled is also listed in Table S1.

Referee 2: Section 3.1: indicate how the filters were rinsed right after the filtration

Reply (P6, L5-7): In this study, filters were not rinsed/washed after the filtration. Therefore, the sentence has been revised as follows:

After filtration, filters were folded without rinsing and wrapped again in aluminium foil and then stored at -20 °C immediately in a freezer onboard before they were brought back to the laboratory for further analysis.

Referee 2: Section 3.2, line 28: detail how the filters were treated with HCI 1N

Reply (P6, L20-22): 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).

Referee 2: Section 3.2, line 30: indicate the diameter of the punches

Reply (P6, L23-25): The diameter of filter has been included in the revised version as follows: Then a half of de-carbonated filter (i.e. a quarter of the original filter - 11 mm) was then punched and placed in tin capsules for further analysis.

Referee 2: Section 3.2, lines 30-31: it looks like $\delta^{15}N$ and PN were analyzed on the decarbonated part of the filter. Why not on the un-decarbonated part of the filter? There is always chance to bias $\delta^{15}N$ and PN using decarbonated material for these measurements (e.g. Lorrain et al (2003) and other references). Also, it looks like there was a very small part of the filters that were analysed for C and N elemental and isotopic composition. What quantities of C and N were analysed?

Reply: We thank the referee to bring the reference Lorrain et al. (2003) to our kind attention.

(P7, L8-18) Lorrain et al. (2003) cautioned that the measurement of PN and δ^{15} N after freezing increases the uncertainty of δ^{15} N and in combination with concentrated HCl treatment, leads to a loss of PN and alteration of the δ^{15} N signature. Therefore, PN content and δ^{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 the present study, the amount of measured C and N ranged from 68.24–322.18 μ gC and 14.46–64.69 μ gN, respectively (Table S1). Precision for δ^{13} C and δ^{15} N decreases for samples containing less than 100 μ gC and 20 μ gN, respectively. Among thirty-six filters analyzed for the present study, only five (three) filters contain less than 100 μ gC (20 μ gN).

Influence of CDW at DCM depth in the ECS

Referee 2: It cannot be stated that the influence of CDW in the study site is nil or insignificant. The low salinity measured at some of the sites (Fig. 3) clearly indicates the influence of CDW. It is mainly the case in surface water but also the case at some of the DCM depths where water was sampled for SPM (stations DH1-1, DH2-1, DH2-2, DH3-1 and CON02). This is also clear from Fig. 6b where five stations falls within the SMW square, SMW being clearly a water body composed of a mixing between KSSW and CDW.

It should better be written that the influence of CDW in the study site is low (see some of the 'minor points' below) or weak (as written in section 5.1, P8, line 34).

Reply: As suggested by the Referee, we have softened the tone of the influence of CDW shown in Fig. 3 and Fig. 6b in the revised text.

End of section 5.1 (P9, lines 1-9) and Fig. 7

Referee 2: Only the DCM depths (= the depths of interest for the present study) should be considered for delineate the polygons of Fig. 7. Was it the case? For similar reason, I think that

the sentence "Interestingly...study area (Fig. 7)" should be deleted or reworded without citing Fig. 7 (but rather Fig. 3?) since it is quite confusing. Another option may be to not cite depth limitation of the water masses influences but only describe Fig. 7.

Reply: We disagree with this suggestion mainly because the conditions of how different water masses (CDW, TWCW and KSSW) are influencing the DCM depths are shown in Fig. 6b. Further, in Fig. 7 we delineated areas influenced by three water masses both horizontally and vertically for the entire water column. Furthermore, the water masses were delineated based on the *T*-S combination and therefore, citing Fig. 3 along with Fig. 7 is fine, but deleting the mention of Fig.7 without water depths may mislead the meaning. Therefore, based on Referee's suggestions, we revised the first paragraph of section 5.1 as follows (**P11, L6-24**):

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 °C, 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 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) indicates the influence of CDW mostly in surface water, but also some of the DCM depths where water was sampled for SPM (Fig. 2). This is also evident from Fig. 6b where these five stations fall within the area of SMW, which is a water body composed of a mixing between CDW and KSSW. However, except 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 water depths by three important water masses: CDW, TWCW and KSSW (Fig. 7). Interestingly, the influence of CDW was constrained only in the upper 0–10 m in five coastal stations during the sampling time, whereas TWCW influences around 0-30 m, covering three-fourths of the study area, and KSSW seems to be largely influenced the bottom water of the entire study area (Figs. 2, 6a and 7).

Referee 2: The last paragraph of section 5.1 is also quite confusing. Reword it as: "In summary, although the river runoff was huge, the influence of CDW plume in the southern part of the ECS was insignificant during summer 2013, mainly because most of the CDW plume was transported to northeastwardly 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 summer 2013."

Reply (P11, L26-31): The last paragraph of section 5.1 has been reworded exactly, as

suggested above by the Referee.

Section 5.2.1

Referee 2: I fully agree the main conclusions of this section and most of data interpretations (especially the first and the last paragraphs).

However the second paragraph adds detailed discussion with literature comparison that is not needed (especially when dealing with zooplankton and Trichodesmium) for the present study. Authors should better goes directly to the conclusion (i.e. the last paragraph) without diluting the main conclusions with unneeded wording. Thus, the second paragraph should be deleted.

Reply (From P12, L23 to P13, L2): As suggested by the Referee, the second paragraph of section 5.2.1 has been deleted in the revised version.

Section 5.2.2

Referee 2: As for the previous section, I fully agree the main conclusions and most of the data interpretation, but this section is too wordy and gives too many details (especially too many values from the literature). Authors should better focus on the main information and the main conclusions.

Thus, I suggest the following:

Referee 2: - P10, lines 26-34: one-two sentence(s) should be enough

Reply (P13, L15-25): Lines 26-34 have been shortened/revised as follows:

Moreover, the POC/Chl *a* ratio of 34.1 g g⁻¹ derived from the slope of a regression line (y = $34.1 (\pm 9.99) \times +49.9 (\pm 8.86)$ (Fig. 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 by Chang et al. (2003) in the ECS and estimated (18–94 g g⁻¹) from phytoplankton cell volumes by the same authors.

Referee 2: - P11, lines 2-4: keep this sentence but rephrase the last line as "filtered particles (Chang et al., 2003; Hung et al., 2013)"

Reply (P13, L25-29): As suggested, the sentence has been rephrased as follows:

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).

Referee 2: - P11, lines 8-10: do not report all these values

Reply (From P13, L33 to P14, L2): These lines have been modified in the revised version as follows:

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 et al. 1996).

Referee 2: Regarding the high POC/Chl a ratio, did authors check if these high values were rather due to very low Chl a concentration or high POC concentration? If the former, these high values may be associated to high uncertainty on the Chl a estimation when values are low. If the latter (high POC concentration associated to Chl a concentration similar to surrounding stations), this may be effectively due to heterotrophic biomass.

Reply: When we use the converted Chl *a* concentration in the revised version, only one SPM shows high POC/Chl *a* ratio of >200 g g⁻¹ (CON02: 303 g g⁻¹). Although it contains neither high POC concentration (CON02: 92.6 μ g L⁻¹) nor high Chl *a* content (CON02: 0.15 μ g L⁻¹), the Chl *a* value seems to be relatively low, as shown in Fig. 8b.

Section 5.3: first three paragraphs

Referee 2: I fully agree the main conclusions and the data interpretations of the first three paragraph of this section.

I suggest authors to have a look at Lowe et al. (2014) and Miller et al (2013): these articles are of interest for the present section.

Reply: These two references are included in the revised version in appropriate places. Please refer to **P15**, **L11-22** in the marked-up manuscript for details.

Page 12, Lines 18-26: two other processes may influence phytoplankton δ^{13} C: temperature and degradation. This is discussed in Savoye et al. (2003) that authors cite in many occurences. Authors may have a look at biplots like δ^{13} C vs temperature and versus POC/Chl *a* and C/N (considering these ratios may also indicate phytoplankton decay). They also may check the normalization of δ^{13} C by temperature (as in Savoye et al., 2003) before plotting normalized δ^{13} C versus POC, since temperature usually have (indirect) influence on phytoplankton δ^{13} C.

Reply (From P17, L24 to P18, L24): As directed, a section 5.4 on "Temperature effect on $\delta^{13}C_{POC}$ data" has been included in the revised version as follows:

5.4 Temperature effect on the $\delta^{13}C_{\text{POC}}$ 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 CO₂, and therefore on phytoplankton δ^{13} C (e.g., Rau et al., 1992; Savoye et al., 2003). The C/N ratio, POC/Chl *a* ratio and δ^{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 δ^{13} C of phytoplankton, we plotted our δ^{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 δ^{13} C of phytoplankton biomass while 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 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, 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: R² = 0.71; p<0.0001; n = 18 and triangles: R² = 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. 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.

Section 5.3: last three paragraphs

Referee 2: I do not think that the last three paragraph of the section are needed. The objective of the two paragraphs before the last (from "The nutrient N/P ratio" to "this mechanism is most likely") is to decipher whether POM sampled in the DCM came from in situ production or from surface production (cf. the fourth paragraph of the section). In fact, these two paragraphs lay on very putative argumentation and do not allow (and are not convincing in) deciphering

between the two hypotheses. These hypotheses have already been discussed in sections 5.2.1 and 5.2.2 with sufficient argumentation for considering that POM mainly came from in situ production. To my point of view, these two paragraphs of section 5.3 are not needed in this section neither in the manuscript.

Reply (From P16, L17 to P17, L13): As suggested, these two paragraphs have been deleted in the revised version.

Referee 2: The last paragraph of the section is a tentative of inventory of POC in the DCM layer. The estimation is very rough, is associated to large uncertainty, and the calculation is not convincing. It is also completely disconnected from the rest of the section, which is focused on δ^{13} C dynamics (see the title of the section). Again, this paragraph is not needed in this section neither in the manuscript. Thus, the last three paragraphs of the section should simply be deleted and the fourth paragraph of the section ("The range... DCM layers?") should be replaced with a brief conclusion of the first three paragraphs of the section.

Reply (P17, L15-21): We agree that the POC inventory in the DCM layer has been done approximately and therefore can be deleted in the revised version.

Referee 2: Section 5.3 would better stand with the first three paragraphs and a conclusion without the unclear and unconvincing last three paragraphs.

Reply: As suggested, all three paragraphs have been deleted in the revised version.

Section 5.4

Referee 2: This section is the less clear and the less convincing of the manuscript. The main conclusion (POM $\delta^{15}N$ distribution is primarily governed by the nutrient status and $\delta^{15}N$ of nitrate) is mainly guess. One of the main issues of the section is the lack of nutrient data. This rend the data interpretation mainly guess-work. Another issue is that authors mainly take into account nitrate as a nutrient for phytoplankton. Ammonium appears only in the last paragraph. The other species of N-nutrient (N₂, dissolved organic nitrogen as urea) are not mentioned. However, it is reported that "Kuroshio Water and TWCW induced Trichodesmium" (P10, line 7). Thus, PN $\delta^{15}N$ values should also be discussed considering N₂-fixers (diazotrophs). At last, many sentences are not clear. This gives the impression that authors do not fully have in mind what processes drives PN and phytoplankton $\delta^{15}N$.

Thus, this section should be deeply reworked including deep data re-interpretation. To me, such section dedicated to PN/phytoplankton $\delta^{15}N$ cannot stand without data of nutrient concentration originating from the same cruise. If these data are not available, PN/phytoplankton $\delta^{15}N$ cannot be discussed. If it would be the case, $\delta^{15}N$ data should be removed from the manuscript.

Reply: Thank you for the suggestion! At present, we don't have depth profiles of nitrate or ammonia or nitrogen isotopic composition of nitrate to strengthen the PN/ δ^{15} N data and related

interpretations. To our knowledge, all our interpretations of PN/ δ^{15} N are based on our understanding of nitrogen dynamics in the ECS and are consistent with the literature cited. This part is also appreciated by Referee 3, but also suggested to strengthen the interpretations using depth profiles of nutrients. However, there is no much information related to δ^{15} N data, especially from the biota-dominated DCM layers, in the East China Sea or South China Sea. Since we proved that the POM is dominated by phytoplankton, publishing δ^{15} N data of this study may fill the data gap and also create some awareness/interests among readers to conduct such investigations in the marginal seas of the western Pacific in detail. Given this, we request anonymous reviewers and Associate Editor to allow this section for publication because we interpreted PN/ δ^{15} N data based on the published information from the East China Sea with some speculations, a practice that is normally encouraged in the scientific field when the availability of data is relatively scarce, such as δ^{15} N of phytoplankton.

Section 5.5

Referee 2: This section gives an important conclusion: the influence of terrestrial POM (mainly originating from the Yangtze River) has drastically decreased in the ECS. This section is mainly based on literature data and conclusions. These inputs from the literature are of interest, but the section should also compare data from the present study with previous data. Thus, this section should start with the comparison of POC/ChI a and C/N ratios, and δ^{13} C values between the present study and previous studies. Then, the decrease of terrestrial POC fluxes and deposition can be cited (literature data). Last lines of the section: there are again unneeded details in these lines. Avoid describing the degradation index but directly give the conclusions of Wu et al (2007b).

Reply (From P20, L1 to P21, L4): As directed, section 5.5 has been revised as follows:

The range of POC/ChI *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 results indicated that POM around the water depths of DCM was largely derived from the synthesis of in situ phytoplankton and the influence of terrestrial OM supplied by the Yangtze River to the southern ECS is low. The 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 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 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: $1.27-8.5 \times 10^{12}$ 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.

English language

Referee 2: The language is usually quite understandable, but there are many errors or mistakes.

Part of them is listed in the 'minor points' below. Nevertheless, the whole manuscript should be deeply checked for English language.

Reply: The whole manuscript has been carefully checked for grammatical errors.

Inconsistencies

Referee 2: There are some inconsistencies between values that are cited in the text and values reported in the tables (see 'minor points' below). Please, check the consistency between all the values reported in the text and tables.

Reply: Cross-checked and corrected.

Abstract, Introduction and Summary and conclusions

Referee 2: Sections 'Abstract' and 'Summary and conclusions' should partly be re-written taking into account the above detailed comments.

Reply: As suggested, these parts have been modified as follows (**Abstract: From P1 to P2**, **17; Summary and conclusions: From P21, L8 to P22, L2**):

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 and surface sediments in the marginal seas. Here we 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 $\delta^{13}C_{POC}$ (-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/ChI 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 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 biomarkers are crucial to revalidate whether or not the POM around the DCM water depths is influenced by terrestrial OM in the river-dominated East China Sea.

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 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 $\delta^{13}C_{POC}$ and POC, and phytoplankton species were the main factors; the former explained ~70% of the variability in $\delta^{13}C_{POC}$, after accounted 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 needs to be substantiated by the nutrient data. Our results show that phytoplankton dynamics drive marine POM composition around DCM layers in the southern East China Sea.

Moreover, phytoplankton in the southern East China Sea contain relatively low δ^{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 mixing models. Therefore, our results with highly variable δ^{13} C_{POC} and δ^{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.

Referee 2: The third objective that appear in section Introduction should be removed (see one of my comments dedicated to section 5.3 $\hat{a} \tilde{A} \tilde{T}$ three last paragraphs $\hat{a} \tilde{A} \tilde{T}$ above).

Reply (P4, L15-16): The third objective in the Introduction part has been deleted in the revision.

Minor points

- P1, Line 20: what do you mean with 'straddling'?

Reply: It means locating or moving around DCM depth intervals.

- P2, line 9 and in the whole manuscript: replace 'endmember' with 'end-member'

Reply: Replaced.

- P3, line 8: remove 'which in turn, the elemental and isotopic compositions of marine productivity' since it is not correct

Reply (P3, L23-24): Deleted.

- P5, line 1: depending what you want to say, add 'by' or 'to' between 'decreased' and '86%'

Reply (P5, L20): As suggested, 'by' is added.

- P5, line 17: replace 'had' with 'have'

Reply (P6, L3): Replaced.

- Last sentence of section 3.1: place this sentence in section 3.2 since it is not sample collection. Rename section 3.2 as 'Determination of SPM concentration and analysis of POC, PN, δ^{13} C and δ^{15} N

Reply (P6, L7-15): The last sentence of section 3.1 has been shifted to section 3.2. As suggested by the Referee, section 3.2 has also been renamed in the revised version.

- P5, line 30: replace 'with' with 'and placed in'

Reply (P6, L24): Replaced.

- Section 3.2, P5-6, sentence "organic carbon and nitrogen ... entering the IRMS": remove the sentence since such level of detail is not needed

Reply P6, L28-30): The sentence has been deleted in the revised version.

- Section 3.2: remove the three last sentences ("Conventional...Sigman et al., 2009" since the first one is unneeded detail and the two last ones do not stand in a section dedicated to methods.

Reply (P7, L4-8): As suggested, the last three sentences in section 3.2 have been deleted in the revised version.

- P6, line 22: add 'usually' between 'profiles of Chl a' and 'show'.

Reply (P8, L28): Added and modified.

- Section 4.1.3: since Fig. 3 illustrates at maximum the first 300m of the water column and since the sampling depth was within this depth interval, please do not describe deeper water, either the reading is quite disturbing. Thus, the temperature ranged between 30 and ca. 15 °C.

Reply (P7, L27-33): We agree with the Referee's view and therefore the paragraph describing the range of temperature has been revised as follows:

Figure 2 illustrates the vertical distributions of temperature and salinity along seven transects across the ECS. In the entire study area, temperature in the 300-m water column varied from 15 °C to 30 °C and distinct water column stratification was evident from the temperature profiles (Fig. 2). The temperature decreases when depth increases and the highest temperature (~30 °C) seen mostly in the surface water and the lowest temperature (5 °C) was observed in stations DH7–8 and DH7–9 at water depths of 850 m and 800 m, respectively (not shown). 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).

- P7, line 16: replace 'increasing' with 'increases'; reword "with the high temperature (>30 °C) spreads widely".

Reply: Replaced. Please see our reply to the previous comment.

- P7, line 29: replace 'insignificant' with 'low'.

Reply (P8, L9): Replaced.

- P8, line 4: '4.4' or '4.5' as reported in table 1?

Reply (P33): The correct value is 4.4 and it is corrected in Table 1.

- P8, line 5: '17.7' or '17.8'?

Reply (P33): The correct value is 17.7 and Table 1 is corrected accordingly.

- P8, lines 7-8: please also indicate where the highest POC and PN concentration were located.

Reply (P10, L19-20): The following sentence has been included in the revised version:

The highest concentrations of POC (263 μ g L⁻¹) and PN (52.8 μ g L⁻¹) are associated with station DH5-1 (Fig. 4).

- P8, line 17: '8.0' or '7.8' as reported in Table 1?

Reply (P33): The correct value is 8.0 and Table 1 is revised accordingly.

- P8, lines 17-18: please also indicate where the highest δ^{13} C values were located.

Reply (P10, L30-31): The following sentence has been included in the revised version:

Consistent to the POC concentration, the highest $\delta^{13}C_{POC}$ value (-18.2 ‰) is also associated with station DH5-1.

- P8, line 21: Fig. 10 is cited before Fig. 5. Check the numbering of the figures.

Reply: Fig. 10 is cited after Fig.5, which was cited just above in the text. We have cross-checked all figure numbers in the revised version.

- P8, line 33: SMW is a water body that is composed of a mixture between two other water masses (CDW and KSSW; Fig. 6). So, do not consider SMW as a water mass and remove it from this list.

Reply (P11, L14): Removed.

- P10, line 25: remove the word 'moderate' since this information is not useful here.

Reply (P13, L13): Deleted.

- P10, line 29: '48' or '52' as reported in Table 2?

Reply (P13, L18): According to Table 2, the value 48 is for the northwestern Pacific and the number 52 is for the western Pacific. As suggested by the referees, Table 2 has been deleted in the revised version.

- P11, line 28: replace 'less' with 'low'; delete "and unrecognized content of terrestrial POM".

Reply (P14, L21-22): Replaced and deleted.

- P12, 2: replace 'to be' with 'would be'.

Reply (P14, L28): Replaced.

- P12, line 8: replace 'more positive' with 'less negative'.

Reply (P14, L34): Replaced.

- P12, line 34: delete 'As for species,'.

Reply (P15, L30): Deleted.

- P13, line 7: delete 'that'.

Reply (P16, L3): Deleted.

- P13, line 8, 9 and 10: replace 'larger' with 'higher'

Reply (P16, L4): Replaced.

- P13, lines 9 and 10: replace 'size species' with 'phytoplankton'

Reply (P16, L5): Replaced.

- P15, line 33: replace 'significantly less' with 'low'.

Reply (P20, L8): Replaced.

- P16, line 12: replace 'proved' with 'illustrate'.

Reply (P20, L23): Replaced.

- Table 1: add POC/Chl a values in this table; indicate in the caption what means 'SD'.

Reply (P33): POC/Chl a values have been included in Table 1 with SD abbreviation.

- Figure 1: indicate KSSW on the figure; be consistent with Fig. 6. Indicate on this figure the location of the stations that appear on Fig. 2 and 3 but were not sampled for SPM in the DCM.

Reply: Fig. 1 shows the simplified current pattern in the ECS, and the center of the upwelling region. As one of the water masses, it is appropriate to show KSSW in Figures 6 and 7 along with other water masses.

Reply: Stations where SPM were not sampled at the deep chlorophyll maximum layer are marked in Fig. 1 in the revised version (red circles in Fig. 1).

Figure 6: the two colours are not distinguishable. Choose other colours. Remove 'from' in the second line of the caption. Add 'were' after 'matters' in the third line of the caption.

Reply: The two colours in Figure 6 are changed. Other corrections are included, as suggested.

Figure 7: replace 'black' with 'grey' in the second line of the caption.

Reply: Replaced.

Figure 8: first line of the caption: it is POC vs. PN and POC vs. Chl a.

Reply: Corrected.

Additional references

Lorrain A., N. Savoye, L. Chauvaud, Y-M. Paulet and N. Naulet, 2003. Decarbonation and preservation method for the analysis of organic C and N contents and stable isotope ratios of low-carbonated suspended particulate materiel. Analytica Chimica Acta, 491, 125-133. Lowe, A. T., A. W. E. Galloway, J. S. Yeung, M. N. Dethier, and D. O. Duggins. 2014. Broad sampling and diverse biomarkers allow characterization of nearshore particulate organic matter. Oikos. 123: 1341–1354, doi:10.1111/oik.01392

Miller, R. J., H. M. Page, and M. A. Brzezinski. 2013. _13C and _15N of particulate organic matter in the Santa Barbara Channel: drivers and implications for trophic inference. Mar. Ecol. Prog. Ser. 474:53-66, doi:10.3354/meps10098

Reply: These additional references are included in the revised version.

Thank you very much.

Anonymous Referee #3

General Comment: This manuscript characterized the bulk and isotopic composition of organic matter collected in DCM layer of the south East China Sea in summer time. The study is well designed and neatly presented. It observed the marine derived material is the dominant organic matter in DCM layer. The influence of the Yangtze River is very limited. Additionally, the nitrogen isotopes elucidated the potential role of N2 fixing in middle shelf where TWCW is dominated and remineralized nutrients plays an important role. However, in this point, the depth profile will be very helpful to strengthen the discussion but it is lack in the manuscript. Furthermore, the influence of lateral transport (cross shelf) better to be considered when the authors estimate the nutrients contributions from different sources and POC inventory.

Reply: Thank you for your positive opinions on the overall work presented here. At present, we don't have depth profiles of nitrate or ammonia or nitrogen isotopic composition of nitrate to strengthen the PN/ δ^{15} N data and related interpretations. However, we reiterate that there is no much information related to δ^{15} N data, especially from the biota-dominated DCM layer in the southern East China Sea. Since we proved that the POM is dominated by phytoplankton, publishing δ^{15} N data of this study may fill the data gap and also create more interests among readers to conduct such investigations in the marginal seas of the western Pacific. Given this, we request anonymous reviewers and Associate Editor to allow this section for publication because we interpreted PN/ δ^{15} N data based on the published information from the southern East China Sea.

Specific comments:

Referee 3: 1) The title better be more specific in study region, such as southern East China Sea

Reply: The title of the manuscript has been modified as follows:

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

Referee 3: 2) Abstract: OK

Reply: Thank you.

Referee 3: 3) Introduction: Better to emphasize the status of DCM in ECS and the potential role in POC inventory estimation and hypothesis

Reply: The text related to the POC inventory has been deleted in the revised version, as suggested by the Referee 2.

Referee 3: 4) Methods P5, the filtration volume was in the range of 0.5-2L, the author used

half filter for POC or PN analysis, Did they have enough material for reliable analysis, especially for nitrogen?

Reply: We have mistakenly mentioned the filtration volume in the original manuscript. Our apologies! We filtered 4.1-19.1 L of water samples for the collection of SPM. The volume of filtration has been changed accordingly in the revised version as follows (**P5, L34 to P6, L3**):

The volume of each water sample was measured by graduated cylinder before filtration. Suspended particles were obtained by filtering 4.1–19.1 L seawater collected at the fluorescence maximum layer through 0.7 μ m/47 mm Whatman Glass Fiber Filters (GF/F), which were wrapped in aluminium foil.

In our study, the amount of measured C and N ranged from 68.24-322.18 μ g and 14.46-64.69 μ g, respectively. Precision for δ^{13} C and δ^{15} N decreases for samples containing less than 100 μ gC and 20 μ gN, respectively. Among thirty-six filters analyzed for the present study, only five (three) filters contain less than 100 μ gC (20 μ gN).

Referee 3: 5) Result and interpretations: The order of hydrographic characteristics can be adjusted as salinity, turbidity and chla

Reply (From P7, L25 to P9, L5): As suggested, we rearranged the order of hydrographic characteristics in the revised version.

6) Discussion: In general, the authors gave proper credit to related work and clearly indicate their own new contribution to the biogeochemical cycles in the study area.

Reply: Thank you.

Some minor suggestions:

Referee 3: a. P13-14, How to use C/P ratio to estimate the Yangtze-sourced nutrients for marine primary productivity, how does the lateral cross shelf transport contribute to the POC inventory?

Reply (From P16 to P17, L21): The text related to the C/P ratio and POC inventory have been deleted in the revised version, as suggested by the Referee 2.

Referee 3: b. P15, L20, this paragraph is a bit speculative and need more straightforward data to support itself, the depth profile data could be bit helpful to elucidate.

Reply: Please refer to our reply to the general comment of Referee 3.

Referee 3: c. Comments on 5.5, why the Yangtze River will play an important role in DCM OM in south ECS, the paragraph of TGD can be moved which seems less related to this topic, also

I am confused about how the author summarized in the abstract" SPM investigated here seems not to be influenced by the terrestrial organic matter supplied by the Yangtze River (Changjiang) in summer 2013, a finding that is contrary to a number of previous studies' conclusion.", which is not convinced in this part.

Reply: The transport pathway of Yangtze River debouched sediments to the Okinawa Trough is one of the unsettled issues in the oceanographic studies of the East China Sea. Recently, Chen et al. (2017) suggested that the Changjiang river plume flows southward when sediments are resuspended along the China coast by cyclonic storms. Most previous studies also had shown that cross-shelf transport of Yangtze-derived sediments to the Okinawa Trough. So, one would expect the influence of Yangtze-derived sediments in the southern ECS during summer.

The part of the sentence "a finding that is contrary to a number of previous studies' conclusion" in the abstract has been deleted in the revised version (**P2, L10-11 in the marked-up manuscript**).

Referee 3: d. The quality of Figure 4 and 5 should be improved. Figure 8b, there is 35 samples summarized but all the other plotted based on 36 samples, why?

Reply: The quality of Figures 4 and 5 has been improved. There is no fluorescence/Chl *a* data for station DH6-1 (see Table S1 in the appendix) and therefore plots with Chl *a* data contain only 35 data points, including Fig. 8b.

Thank you very much.

Additional References

- Aminot, A. and Rey, F.: Standard procedure for the determination of chlorophyll *a* by spectroscopic methods, ICES Techniques in Marine Environmental Sciences, Copenhagen, Denmark, 8–11, 2000.
- Cullen, J. J., Reid, F. M. H., and Stewart, E.: Phytoplankton in the surface and chlorophyll maximum off southern California in August, 1978, J. Plank. Res., 4, 665–694, 1982.
- Fontugne, M. R.: Les isotopes stables du carbone organique dans l'océan: application à la paléoclimatologie, PhD thesis, Université de Paris XI, 1983.
- Fontugne, M. R. and Duplessy, J. -C.: Organic carbon isotopic fractionation by marine plankton in the temperature range –1 to 31°C, Oceanol Acta, 4, 85–90, 1981.
- Lorrain A., Savoye, N., Chauvaud, L., Paulet Y. -M., and Naulet, N.: Decarbonation and preservation method for the analysis of organic C and N contents and stable isotope ratios of low-carbonated suspended particulate material. Anal. Chim. Acta, 491, 125–133, 2003.
- Lorenzen, C. J.: Determination of chlorophyll and pheo-pigments: Spectrophotometric equations, Limnol. Oceangr., 12, 343–346, 1967.
- Lowe, A. T., Galloway, A. W. E., Yeung, J. S., Dethier, M. N., and Duggins, D. O.: Broad sampling and diverse biomarkers allow characterization of nearshore particulate organic matter, Oikos, 123, 1341–1354, 2014.
- Malone, T. C., Falkowski, P. G., Hopkins, T. S., Rowe, G. T., and Whitledge, T. E.: Mesoscale response of diatom populations to a wind event in the plume of the Hudson River, Deep-Sea Res., 30, 149–170, 1983.
- Miller, R. J., Page, H. M., and Brzezinski, M. A.: δ¹³C and δ¹⁵N of particulate organic matter in the Santa Barbara Channel: drivers and implications for trophic inference, Mar. Ecol. Prog. Ser., 474, 53–66, 2013.
- Rau, G. H., Takahashi, T., Des Marais, D. J., Repeta, D. J., and Martin, H.: The relationship between δ¹³C of organic matter and [CO₂(aq)] in ocean surface water: data from a JGOFS site in the northeast Atlantic Ocean and a model, Geochim. Cosmochim. Acta, 56, 1413– 1419, 1992.
- Sackett, W. M., Eckelmann, W. R., Bender, M. L., and Bé, A. W. H.: Temperature dependence of carbon isotope composition in marine plankton and sediments, Science, 148, 235–237, 1965.
- Wong, W. W. and Sackett, W. M.: Fractionation of stable carbon isotopes by marine phytoplankton, Geochim. Cosmochim. Acta 42, 1809–1815, 1978.

Biogeochemical characteristics of suspended particulates at deep chlorophyll maximum layers in the <u>southern</u> East China Sea

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- 15 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 <u>thus</u> frequently used <u>to for</u> characteriz<u>e and distinguishing and its their</u> sources in suspended particulates and surface sediments in the marginal seas. Here we investigate<u>d</u> suspended particulate matters (SPM) collected <u>around from the</u> deep
- 20 chlorophyll maximum (DCM) layers in the continental shelf of the southern East China Sea for particulate organic carbon and nitrogen (POC and PN) contents and their isotopic compositions (δ¹³C_{POC} and δ¹⁵N_{PN}) to understand the provenance and dynamics POMbiogeochemical characteristics of POM straddling at biotic dominated DCM depths. 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
- 25 showed a large variation in $\delta^{13}C_{POC}$ (-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
- 30 seawater, as indicated by similar spatial distribution between δ¹⁵N_{PN} and the current pattern and water masses in the East China Sea; although interpretations of δ¹⁵N_{PN} data should be verified with the nutrient data in future studies. When combined with hydrographic parameters, such as temperature, salinity and turbidity, and

chlorophyll *a* (Chl *a*), these elemental and isotopic results revealed that POM in the DCM layers was largely from the newly-produced, *in situ* phytoplankton-dominated OM and have wider $\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$ -compositions than previously thought. As supported by the POC to Chl *a* ratio, a large variation of $\delta^{13}C_{POC}$ was resulted from the changes in primary productivity and phytoplankton species, whereas the nutrient status and $\delta^{15}N$ of dissolved

- 5 nitrate were the main controlling factors of δ¹⁵N_{PN} variability in the DCM layers. Consistently, the spatial distribution of δ¹⁵N_{PN} showed a similarity with the current pattern in the East China Sea, with ¹⁵N-enriched freshwater in the coastal region and Kuroshio Water in the northeast of Taiwan Island, but nutrient-depleted Taiwan Warm Current Water in the mid-shelf; as the latter seems to have promoted the N₂-fixation, resulting in the depleted δ¹⁵N_{PN}-in the mid-shelf. Furthermore, SPM investigated was here seems not to be weakly influenced
- 10 by the terrestrial <u>OMorganic matter</u> supplied by the Yangtze River (<u>Changjiang</u>) <u>during in-summer 2013</u>, a finding that is contrary to a number of previous studies' conclusion <u>due to the reduced sediment supply by the Yangtze River and north-eastward transport of riverine particles to the northern East China Sea</u>. 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, given the complications associated with stable isotopes of organic
- 15 matter, additional parameters such as <u>data of</u> radiocarbon and biomarkers are crucial to revalidate whether or not <u>the POM aroundSPM in</u> the DCM <u>water</u> depths is influenced by terrestrial <u>OM organic compounds</u> in the riverdominated East China Sea.

1 Introduction

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Stable isotopes of organic carbon and nitrogen (δ¹³C, δ¹⁵N) and molar carbon to nitrogen (C/N) ratios are the most frequently used natural tracers to identify for identifying 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 δ¹³C, δ¹⁵N and C/N ratios between different endmembers, especially 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 of end-members by using mass balance models thus requires known and constant values of elemental and isotopic end-members of major sources of OM to the depositional system (e.g., Goñi et al., 2003). Any study applying mixing models for the OM source discrimination should therefore clearly

30 identify representative values for the local sources of OM inputs into the area under investigation. However, in most cases, end_member values of δ¹³C, δ¹⁵N and molar C/N ratios were simply replaced by 'typical' numbers, such as ca. –20 ‰ and –27 ‰ for δ¹³C of marine phytoplankton and terrestrial plants, respectively, but without measuring 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 end_member mixing

models to distinguish marine versus terrestrial 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

- 5 representative of these values in primary production-derived OM and in that they are largely synthesized by phytoplankton (Gearing et al., 1984). Since phytoplankton is the main primary producer of marine OM, Phytoplankton as the primary producer of marine OM-the elemental and isotopic compositions of phytoplankton should therefore be considered while when studying the dynamics of POM in the marine water column.
- 10 It is known that stable isotopes (δ¹³C, δ¹⁵N) and molar C/N ratios of POM in estuarine and marine areas are representative of these values in primary production derived OM and in that they are largely synthesized by phytoplankton (Gearing et al., 1984). The cChlorophyll *a* (Chl *a*) concentration in the sea water is often used as an index of phytoplankton biomass and phytoplankton carbon (Cullen et al., 1982; Malone et al., 1983)., and t The deep chlorophyll maximum (DCM) layer, which contributes significantly have a significant contribution to the
- 15 total biomass and primary production in the whole water column (Weston et 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 <u>layer</u> has been explained by several mechanisms: the differential zooplankton grazing with depths (Riley et al., 1949; Lorenzen, 1967), adaption of the phytoplankton to light intensities or to increased concentration of nutrients (Nielsen and 2007).
- Hansen, 1959; Gieskes et al., 1978), chlorophyll accumulation by sinking <u>detritus</u> of phytoplankton <u>detritus</u> (Gieskes et al., 1978; Karlson et al., 1996), and <u>the</u>-decomposition of chlorophyll by light (Nielsen and Hansen, 1959). <u>The</u> DCM <u>layer</u> is common in both coastal and open oceans, occurring at <u>a</u>-relatively shallow depths (1–50 m) in coastal seas, but in deeper depths (80–130 m) in open ocean (<u>Cullen, 1982;</u> Gong et al., 2015), and often variable in time and space (Karlson et al., 1996). For example, the 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, δ¹³C, δ¹⁵N and molar C/N ratios of POM in <u>the</u> DCM layers <u>of in</u> the continental shelf waters should reflect the δ¹³C, δ¹⁵N and molar C/N ratios of phytoplankton, which in turn, the elemental and isotopic compositions of marine productivity (Savoye et al., 2003; 2012; Gao et al., 2014).
- 30 East China Sea is, one of the largest marginal seas in the world, receivinges huge quantities of freshwater (905.1 km³ yr⁻¹; Dai et al., 2010) and organic carbon (2.93 Tg C yr⁻¹, Tg = 10¹² g; Qi et al., 2014) from the Yangtze River (Changjiang). Nutrient-richenriched freshwater input in turn stimulates the water column productivity significantly in coastal waters compared to the open ocean. The annual primary production for the entire shelf of East China Sea is high among the marginal seas and has been estimated to be 85 Tg C yr⁻¹ in 2008 (Tan et al., 2011).

Several studies have been carried out on the physical, chemical and biological aspects of East China Sea, including distributions of seasonal currents (e.g., Gong et al., 2010), chemical hydrography and nutrients distribution (Chen, 1996, 2008) and phytoplankton species in the water column (e.g., Zheng et al., 2015; Jiang et al., 2015). Likewise, δ^{13} C, δ^{15} N and molar C/N ratios of POM have been constrained in a limited number of

- 5 transects across the East China Sea (e.g., Wu et al., 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 East China Sea, especially along the indirect transport pathway of the Yangtze-derived terrestrial material to the Okinawa Trough (Chen et al., 2017), are almost unavailable. In a recent study, Gao et al. (2014) investigated elemental and isotopic compositions of POM
- 10 these parameters in surface, DCM and bottom layers waters in different seasons and years, but they focused on the northern part of the East China Sea with a scanty attention has been paid on the biogeochemical processes involved in the DCM layers. Here, we investigate δ¹³C, δ¹⁵N and molar C/N ratios of suspended POM in DCM layers 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 of East China Sea; (2) to understand the factors
- 15 controlling δ^{13} C and δ^{15} N <u>dynamics</u> in DCM layers <u>of the southern East China Sea</u>; and (3) to estimate the POC inventory in DCM layers of <u>the East China Sea</u>.

2 Study area

The East China Sea (ECS; Fig. 1) is the largest river-dominated marginal sea in the northwestern Pacific region. The continental shelf of <u>the</u>ECS is relatively shallow (<130 m) with an average water depth of 60 m, but wide (>500 km). The Yangtze River (Fig. 1), with a catchment area of more than 1.94 × 10⁶ km² (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 km³ 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 × 10⁹ mol yr⁻¹ for the interval of 1981–2006; Chai et al., 2009, and references therein) and sediments from the Yangtze River, the ECS is characterized by the complex circulation pattern <u>that is</u> largely driven by the seasonally reversing East Asian monsoon winds (He et al., 2014; Chen et al., 2017). The surface circulation in the shelf of ECS is characterized by the south-north China Coastal Current (CCC) in the west, northward-moving Taiwan Warm Current (TWC) in the middle part and the north-northeastward-flowing Kuroshio Current (KC) in the east (Fig. 1) (Liu et al., 2006). <u>The</u> Changjiang Diluted Water (CDW) is a mixture of freshwater of Yangtze River and <u>the</u> shelf water of East China Sea, characterized by a low salinity (<30, Umezawa et al., 2014). Owing to <u>a</u> huge <u>amount of</u> freshwater

<u>discharge input from of the Yangtze into the ECS</u>, it has been believed that the CDW is the main source 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

- 5 al., 2004). 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) in the northeast off Taiwan Island due to an abrupt change of seafloor topography in the outer shelf of <u>the ECS</u> (dashed ellipse in Fig. 1) (Su et al., 1989; Sheu et al., 1999). The upwelled, oxygen-unsaturated KSSW is characterized
- 10 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 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 made up 90% of the shelf water in the ECS (Chen, 1996; Sheu et al., 1999).
- 15 The primary productivity in the ECS is limited by nitrogen deficiency in summer, but light in winter (Chen et al., 2001; Chen and Chen 2003). With the highest primary production during summer, It -annual primary production showed distinct spatial and temporal variations, with the highest primary production in summer and an annual primary production 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 ECS, south- eastern ECS and the entire ECS, respectively, in 1998 (Gong et al., 2003). The primary productivity has however
- 20 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 Material and methods

25 **3.1 Sample collection**

To investigate the biogeochemical characteristics of POM in DCM layers of <u>the southern</u> East China Sea, suspended particles <u>around the DCM water depths (10–130 m)</u> were collected <u>at around water depths of DCM</u> from thirty-six stations <u>along in</u>-seven transects across the continental shelf by the *Science 3* cruise (organized by the Institute of Oceanography, Chinese Academy of Sciences) during <u>summer (June 22–July 21)</u>, 2013 (Fig. 1). At each site, the physical properties of <u>the</u> 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 PVC bottles. The volume of each water

sample was measured by graduated cylinder before filtration. Suspended particles were obtained by filtering 0.5-2-4.1-19.1 L seawater collected at the fluorescence maximum layer through 0.7 µm/47 mm Whatman Glass Fiber Filters (GF/F), which were wrapped in aluminium foil. All GF/F filters <u>have had</u> been pre-combusted at 450 ^oC for 4 h in a muffle furnace to remove the background carbon and pre-weighed for determining the concentration of suspended particulate matters (SPM). After filtration, filters were folded <u>without rinsing</u> and wrapped again in aluminium foil and <u>then</u> stored at -20 °C immediately in a freezer onboard before they were brought back to the laboratory for further analysis. In the laboratory, filters with suspended particles were freezedried and filters for SPM concentration were further dried in an oven at 50 °C for 48 h. The weight difference between the dried filter and its counterpart before filtration was used to calculate the weight of SPM.

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3.2 Determination of SPM concentration and Aanalyseis of ChI 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 its counterpart before the filtration was used to calculate the

- 15 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).
- 20 Prior to the measurement of POC and PN contents and their stable isotope values (δ¹³C_{POC} and δ¹⁵N_{PN}) in SPM samples, a half of each filter was <u>placed in a culture dish and treated with 1N HCl 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). Decarbonated sample was dried at 50 °C for 48 h in an oven for HCl evaporation. Then <u>a</u> half of <u>the</u> de-carbonated filter (i.e. a quarter of the original GF/F filter, ~11 mm) was then punched <u>and placed in with tin capsules</u> for further analysis. The POC and PN contents and their δ¹³C_{POC} and δ¹⁵N_{PN} compositions were measured at the</u>
- Stable Isotope Facility of University of California Davis in USA, by using an elemental analyzer (EA) (Elementar Analysensysteme GmbH, Hanau, Germany) interfaced to a continuous flow isotope ratio mass spectrometer (IRMS; PDZ Europa 20–20, Sercon Ltd., Cheshire, UK). Organic carbon and nitrogen in samples were combusted at 1080 °C in a reactor and transformed into CO₂ and N₂, respectively, which were then separated
- 30 through a molecular sieve adsorption trap before entering the IRMS. During the isotopes (δ¹³C_{POC} and δ¹⁵N_{PN}) analyses, different working standards (Bovine Liver, Glutamic Acid, Enriched Alanine and Nylon 6) of compositionally similar to the samples being analyzed 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 0.2 ‰ for δ¹³C and 0.3 ‰ for δ¹⁵N. 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 N₂ for nitrogen, that is δX (‰) = [(R_{sample} - R_{standard})/R_{standard}] × 10³, where X = ¹³C or ¹⁵N, R = ¹³C/¹²C or ¹⁵N/¹⁴N, R_{sample} and R_{standard} are the heavy (¹³C or ¹⁵N) to light (¹²C or ¹⁴N) isotope ratios of sample and standard, respectively (e.g., Selvaraj et al., 2015). Conventional standards have an isotope value of 0 ‰ for δ^{13} C of VPDB (a marine limestone) and δ^{45} N of N₂ in air, respectively. In general, organic carbon synthesized by biota is usually depleted

5 limestone) and δ¹⁵N of N₂ in air, respectively. In general, organic carbon synthesized by biota is usually depleted in ¹³C relative to VPDB and has a negative δ¹³C value (Fry and Sherr, 1989). On the other hand, δ¹⁵N values of POM do not show a routine mainly due to their enriched oxidation states (Sigman et al., 2009).

Lorrain et al. (2003) cautioned that the measurement of PN and δ¹⁵N after freezing increases the uncertainty of
 δ¹⁵N and in combination with the concentrated HCl treatment, leads to a loss of PN and alteration of the δ¹⁵N signature. Therefore, PN content and δ¹⁵N 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 δ¹⁵N values (~3.8–

15 <u>8.4 ‰) obtained in the present study is comparable to the range of δ¹⁵N values (ca. 0.7–9.4 ‰) obtained by Wu et al. (2003) for the entire water column. In addition, precision for δ¹³C and δ¹⁵N 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).</u>

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4 Results and interpretations

4.1 Hydrographic characteristics and chlorophyll a

25 <u>4.1.1 Temperature and salinity</u>

Figure 2 illustrates the vertical distributions of temperature and salinity along seven transects across the ECS. In the entire study area, temperature in the 300-m water column varied from 15 °C to 30 °C and distinct water column stratification was evident from the temperature profiles (Fig. 2). The temperature decreases when depth increases and the highest temperature (~30 °C) seen mostly in the surface water and the lowest temperature (5 °C) was observed in stations DH7–8 and DH7–9 at water depths of 850 m and 800 m, respectively (Fig. 2). 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 °C, indicating the limited influence of CDW

- 5 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</p>
- 10 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 a low turbidity (<2.0 FTU), except for stations CON02 (4.75),</p>

20 <u>DH5–1 (3.44), and DH7–1 (5.52) (Fig. 3).</u>

4.1.<u>3</u>4. Chlorophyll <u>fluorescence and chlorophyll</u> a (Chl a)

- The concentration of chlorophyll fluorescence concentration varied up to18.0 µg L⁻¹ in the study area (Fig. 2<u>3</u>). The highest Chl fluorescence concentration was observed in surface water atef station DH3–1, and all other remaining values are less than 8.0 µg L⁻¹ (Fig. 3). The vertical profiles of Chl fluorescence usually show a clear maximum in the subsurface layer at around 20 m in near coastal stations and 50 m in outer shelf stations (Fig. 2<u>3</u>). 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 layers and/or contiguous to the DCM layer (open squares in Fig. 2<u>3</u>), ideally representing the biogeochemical behaviours of POM straddling in DCM layers. 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
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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.

<u>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 $R^2 = 0.93$ (Fig. S1 in the Supplementary material). This relationship was used to convert the fluorescence values to Chl *a* concentrations of all the remaining SPM.</u>

10 using an equation: $y = 0.708 \times + 0.199$, where y is Chl *a* and x is chlorophyll fluorescence concentration. 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 converted Chl *a* values were used to calculate POC/Chl *a* ratio (Table S1), which is discussed in section 5.2.2.

15 4.1.2 Turbidity

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The turbidity in the water column of the ECS varied from 0.0 to 20.9 Formazin Turbidity Unit (FTU) (Fig. 2). 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. 2). This indicates sediment resuspension from the sea floor, which was probably induced by hydrodynamic forces such as tides, waves and currents in the shallow

- 20 floor, which 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 from SPM sampled have a low turbidity (<2.0 FTU), except for stations CON02 (4.75), DH5–1 (3.44), and DH7–1 (5.52) (Fig. 2).</p>
- 25 4.1.3 Temperature and salinity

Figure 3 illustrates the vertical distributions of temperature and salinity along seven transects in the ECS. In the entire study area, temperature in the water column varied from 5 °C to 30 °C and distinct water column stratification was evident from the temperature profiles (Fig. 3). The temperature decreases when depth increasing, with the high temperature (>30 °C) spreads widely in the surface water and the lowest temperature (5 °C) was observed in stations DH7 8 and DH7 9 at water depths of 850 m and 800 m, respectively (Fig. 3). The variation of temperature at SPM sampling depths ranges from 19.1 °C to 28.2 °C, showing a general decreasing trend from the inner to outer shelf in each transect (Fig. 3).

Salinity shows a general increasing trend with water depths (Fig. 3), varying from 26.9 to 34.8 with an average value of 34.6 in the entire water column. An increasing trend of salinity from the west to east in all seven transects can be found (Fig. 3). The low salinity (<30) was constrained in the upper 10 m in four coastal stations (DH1–1, DH2–1, DH3–1, CON02; Fig. 3), wherein temperature is <24 °C, indicating the limited influence of CDW

- 5 plume. 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. 3), 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. 3) and varies from 32.7 to 34.7 with an average salinity of 34.0, indicating insignificant influence of CDW at DCM
- 10 depths in the ECS.

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 higher values in the inner shelf region and lower values in the outer shelf region (Fig. 4), consistent with the water column turbidity (Fig. 23). The POC Concentration of POC 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 resembles each other (Fig. 4). The highest concentrations of POC (263 µg L⁻¹) and PN (52.8 µg L⁻¹) are associated with station DH5-1 (Fig. 4 and Table S1). Higher concentrations of POC (>90 µg L⁻¹) and PN (>21 µg L⁻¹) are mostly observed in the inner shelf along the coastal line, decreasing gradually towards the offshore direction (Fig. 4). The lowest concentrations of POC and PN are observed in the easternmost stations, nearby off northeast Taiwan Island (Fig. 4). Although the concentrations of both POC and PN varied more than an order of magnitude (Fig. 4), the molar C/N ratios are fairly uniform at DCM layers of the entire ECS, ranging from 4.1 to 6.3 with a mean ratio of 5.6±0.5 (n = 36) (Table 1).

4.3 δ¹³C_{POC} and δ¹⁵N_{PN}

Spatial distributions of δ¹³C_{POC} and δ¹⁵N_{PN} in <u>SPM</u> at around DCM layers are presented in Fig. 5. δ¹³C_{POC} decreased from the inner shelf to offshore region, varying widely from –25.8 ‰ to –18.2 ‰ (Table 1). <u>Consistent</u> to the POC concentration, the highest δ¹³C_{POC} value (–18.2 ‰) is also associated with station DH5-1. The range of δ¹⁵N_{PN} is 4.2 ‰, varying between 3.8 ‰ and 8.0 ‰ (Table 1). The lowest δ¹³C_{POC} values (–25.8 ‰ and – 25.2 ‰) are found in the Okinawa Trough, off northeast Taiwan Island, while the δ¹⁵N_{PN} values in the same locations are also relatively higher (6.73 ‰ and 7.78 ‰) than that of the surrounding location the nearby locations (Fig. 5). The spatial distribution of $\delta^{13}C_{POC}$ is quite similar to the spatial distribution of POC (Fig. 4), and the correlation coefficient (R²) between $\delta^{13}C_{POC}$ and POC was 0.55 (p<0.0001; Fig. 10).

5 Discussion

5

5.1 Influences of different from 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 at around DCM layers (Fig. 6b). The T-S diagram for all the water depths (Fig. 6a) shows a convergence region at around 17 °C, 34.6 (Fig. 6a), 10 representing the upwelling of KSSW (Umezawa et al., 2014). There are two trends in the T-S diagram, indicating a mixing of three 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 is mixed primarily by three four water masses, CDW, SMW, KSSW, and TWCW (Fig. 6a). The low salinity observed at five coastal sites (DH1-1, DH2-1, DH2-2, DH3-1 and CON02; Fig. 15 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 also evident from Fig. 6b where five stations fall within the area of SMW, which is a water body composed of a mixing between CDW and KSSW. However, except these five coastal stations, the most DCM water at depths where from water was sampled for SPM sampled seems to be weakly influenced by the CDW (Fig. 6b). Furthermore, bBased on the T-S range of different water masses (Fig. 6), we further 20 delineated the area influenced along with water depths byef three important water masses: CDW, TWCW and KSSW (Fig. 7). Interestingly, the influence of CDW was constrained only in the upper 0-10 m in five four coastal stations during the sampling time, whereas TWCW influences around 0-30 m, covering three-fourths of the study area, and KSSW seems to be largely influenced the bottom water of the entire study area (Figs. 2, 6a and 7).

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

5.2 Characterization of POM in DCM layers

35 5.2.1 Molar C/N Ratio

A necessary first step in the source analysis of POM using bulk carbon and nitrogen isotopes as well as the molar carbon <u>to and</u>-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

- 5 (R² = 0.98, p<0.0001; Fig. 8a) between POC and PN (R² = 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 representing 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 of ECS are lower than the canonical Redfield ratio</p>
- 10 (6.63) (Fig. 8a), but are similar to the average 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 <u>it</u> is higher than an unprecedented low C/N ratio (2.65±0.19) of POM in Canada Basin<u>that</u>, which was attributed to a dominant contribution of smaller sizes (<8 µm) phytoplankton to POC (Crawford et al., 2015). Wu et al. (2003)</p>
- 15 investigated the C/N ratio of POM at all depths <u>along in</u> the *PN* transect in ECS (4.3–29.2), 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 <u>a wider that the C/N ratio ranged</u> 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 in-1994. The authors attributed the lower C/N in spring to an intense biological activity than in autumn, and the spatial distribution of
- 20 attributed the lower C/N in spring to an intense biological activity C/N was thought to be related to that of phytoplankton abundance.

Comparing with C/N ratios of POM in Wu et al. (2003) and Liu et al. (1998), the C/N ratio at DCM layers in this study is relatively uniform and low. Such distinct characteristic of low C/N ratio in DCM layers indicate a protein-

25 enriched chemical composition in POM. As the Redfield ratio is achieved when OM contains about 45 % protein (C/N = 3.8) and 10 % nucleic acids (C/N = 2.6) (Geider and La Roche, 2002), the relatively low C/N ratio in DCM layers is consistent with the C/N ratio of phytoplankton (<50 μm) (4.5 5.9) in surface water and zooplankton (200-363 μm) (5.3-6.4) from 10 m above the bottom to surface water of the northern ECS in July 2010 (Chang et al., 2014). In addition, Kuroshio Water and TWCW induced *Trichodesmium* (Chen et al., 1996b; Jiang et al., 2017)

- 30 may be partly responsible for the low C/N ratio in POM at DCM layers. According to Mague et al. (1977) and Letelier and Karl (1996, 1998), the C/N ratio of *Trichodesmium* varied narrowly from 4.1 to 7.3, and the relatively low C/N ratio in *Trichodesmium* was thought to be due to its nitrogen fixing capability (Geider and La Roche, 2002; Chen et al., 2004). Moreover, the inshore offshore decreasing trend of abundance of water mass related *Trichodesmium* in ECS during summer (Jiang et al., 2017) is consistent with the general inshore offshore
 - 12

increasing distribution of δ^{15} N in POM at DCM layers in this study, except for two stations in the northeast of Taiwan Island (Fig. 5), which will be discussed later.

Characteristically, a narrow range of low C/N ratios in our SPM samples confirms the lack of terrestrial signals

- 5 transported mainly by the Yangtze River. We therefore suggest that the POM in <u>the DCM</u> layers of East China Sea is dominated by marine-sourced OM with an unrecognized contribution of terrestrial OM. <u>The IL</u>ow C/N ratios further restrict the assumption of degradation of nitrogen-<u>richenriched</u> OM, <u>a process that which</u> normally increases the C/N ratio than <u>that of</u> the Redfield ratio. Therefore, the molar C/N ratio can be better explained as a source signal of OM rather than OM degradation in <u>the</u> SPM <u>samples</u>-investigated in this study.
- 10

5.2.2 POC/Chl a Ratio

The moderate-linear correlation ($\mathbb{R}^2 = 0.49$, p<0.0001; Fig. 8b) between POC and Chl a ($\mathbb{R}^2 = 0.49$, 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 35.3_34.1 g g⁻¹ derived from the slope of a regression line (y = 34.1)

- 15 samples. Moreover, the POC/Chl *a* ratio of 35.3 34.1 g g⁻¹ derived from the slope of a regression line (y = 34.1 (±9.99) x +49.9 (±8.86) 35.3 (±8.56) x + 44.0 (±6.27) (Fig. 8b) is consistent with the reported POC/Chl *a* ratios in the ECS (36.1 g g⁻¹; of 36.1 g g⁻¹ for SPM samples at 40 m water depths in the ECS (Chang et al., 2003) and the North-western Pacific (48 g g⁻¹; and nearly similar to the reported POC/Chl *a* ratio of 48 g g⁻¹ for the DCM layer of Northwestern Pacific (Furuya, 1990) (Table 2). However, the POC/Chl *a* ratio obtained in this study is lower than
- 20 that estimated (64 g g⁻¹) for the sinking particles at around 20 m in the inner shelf of in the ECS and 100 m at the Kuroshio region, off northeast Taiwan Island (Hung et al., 2013) (Table 2). The range is well within the range (13–93 g g⁻¹) reported for POM in the ECS based on the similar regression analysis between POC and Chl a concentration by Chang et al. (2003) for POM at 2 m water depth in the ECS and , which is also consistent with the range (18–94 g g⁻¹) for phytoplankton carbon to Chl a ratio, estimated from phytoplankton cell volumes by
- 25 the same authors (Table 2). 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 by acetone with (Chang et al., 2003;) or without acidification (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 close to 40 and 140 g g⁻¹ (Geider, 1987; Thompson et al. 1992; Montagnes et al.

1994<u>; Head et al. 1996</u>), lower than 70 g g⁻¹ (Geider, 1987), lower than 100 g g⁻¹ (Head et al. 1996) or lower than 140 g g⁻¹ (Thompson et al. 1992). 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)

- 5 (Cifuentes et al., 1988; Savoye et al., 2003; Liénart et al., 2016, 2017). The POC/Chl *a* ratio in the DCM layer of East China Sea is almost <200 g g⁻¹ (<u>3326</u>-200 g g⁻¹), with <u>one exception (CON02: 303 g g⁻¹two exceptions (DH5-2: 369 g g⁻¹; CON02: 617 g g⁻¹</u>; Fig. 9), indicating that POM in <u>the DCM layers of the ECS was dominated by phytoplankton</u>, as also indicated by the low C/N ratios (4.1–6.3). The relatively high POC/Chl *a* ratios only in <u>one station</u>, two stations, DH5-2 and CON02 (Fig. 9), suggest that <u>the POM in this sample these two samples</u>
- 10 was likely <u>sourced</u> from degraded phytoplankton OM, terrestrial OM, or heterotrophic-dominated OM. However, the molar C/N ratio of DH5-2 (5.6) and CON02 (5.3) are 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. 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-
- 15 <u>richenriched</u> 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 <u>CON02</u> these two stations seems to be dominated by heterotrophic biota, though the exact reason for the dominance of heterotrophic biota only at <u>one location</u> two locations in our study area is unknown and needs further investigation.
- 20 Briefly, several clues indicate the predominance of newly-produced, phytoplankton-synthesized OM in the DCM layers of <u>the southern</u> East China Sea: 1) <u>low less</u> influence of fresh water-<u>and unrecognized content of terrestrial POM</u>, 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.

25 5.3 Dynamics of $\delta^{13}C_{POC}$ in POM in DCM

Although a narrow range of molar C/N ratio in <u>the SPM indicatedsuggested</u> an aquatic origin for <u>the POM at DCM</u> layers, the wide variability of δ¹³C_{POC} (-25.8 to -18.2 ‰) suggests that <u>the POM at DCM</u> layers to would be a mixture of terrestrial C3 plants with a typical δ¹³C value of ca. -27 ‰ (e.g., Peters et al., 1978; Wada et al., 1987)
and marine phytoplankton with a typical δ¹³C range of -18 to -20 ‰ (e.g., Goericke and Fry, 1994). However, Fig. 5 illustrates a distinct decreasing trend of δ¹³C_{POC} towards the outer shelf; a pattern opposite to an increasing trend of δ¹³C 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; Kao et al., 2003; Wu et al., 2003). Such a spatial distribution with more positive less negative δ¹³C_{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), <u>areis</u> 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 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 northeastern Taiwan, likely <u>due to influenced by</u> the intrusion of Kuroshio Subsurface Water with low $\delta^{13}C$ from – 31 ‰ to -27 ‰ (Wu et al., 2003), <u>is as also in agreement</u> with the hydrographic parameters of this location (Figs. 3-2 and 7).

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A positive linear correlation between δ¹³C_{POC} and POC and δ¹³C_{POC} (R² = 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 CO₂, 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 ¹³C-enriched OM in phytoplankton (Fry and Wainwright, 1991; Nakatsuka et al., 1992; Miller et al., 2013). Lowe et al. (2014) observed increased δ¹³C 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 δ¹³C_{POC}, only 55 % of δ¹³C_{POC}
variation can be explained by primary productivity (Fig. 10a), implying that other factors, such as species and sizes of phytoplankton, must have influenced δ¹³C values of in-phytoplankton living in the DCM layers.

In East China Sea, tThe 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 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, which is widely believed to be due to nutrient availability (Zheng et al., 2015). As for species, tThe phytoplankton species have distinct spatial characteristics, but no significant differences in species differences 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)

 μ m) to Chl *a*, respectively, was 40 %, 46 % and 14 % in <u>nutrient-rich</u> inshore <u>nutrient-rich</u> 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 that transported by Taiwan Warm Current and Kuroshio Current. It has been reported that diatoms have <u>higher larger</u> δ¹³C values (–19 to –15 ‰) than dinoflagellates (–22 to –20 ‰;

- 5 Fry and Wainwright, 1991; Lowe et al., 2014). Likewise, large phytoplankton size species have larger higher δ¹³C values than small phytoplankton size species and heterotrophic dinoflagellates have higher large δ¹³C values than autotrophic dinoflagellates (Kopczyńska et al., 1995). Similarly, wide variations of δ¹³C_{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 δ¹³C in suspended OM that was dominated by phytoplankton were reported from in-the Delaware
- 10 estuary (-25 to -20 ‰; Cifuentes et al., 1988), the Bay of Seine (-24.3 to -19.7 ‰; Savoye et al., 2003) and 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)was reported, which was dominated by phytoplankton. 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).

The range and distribution of δ¹³C_{POC} in newly produced POM has been comprehensively deciphered by variations in primary productivity (or biomass) and phytoplankton species compositions in East China Sea, as discussed above. However, it remains unclear that is this newly produced POM in DCM made by phytoplankton

- 20 in surface water that subsequently sank to DCM depths or made by *in situ* phytoplankton, which inhabit in the DCM layers?The nutrient N/P ratio and a selective zooplankton grazing are mostly controlling the distribution of phytoplankton community in the East China Sea (Guo et al., 2014). The zooplankton grazing is important for phytoplankton species abundance when a selective grazing on one specific species. Pilati and Wurtsbaugh (2003) demonstrated that zooplankton grazing coupled with nutrient transport is of importance for the persistence of
- 25 DCM in a nutrient depleted mountain lake. The DCM depths in the East China Sea were near the nitracline and an upward vertical transport of nutrients from the sea bottom can contribute significantly to the DCM layer and maintain the chlorophyll maximum in the shelf region of ECS (Lee et al., 2016). Based on the nutrient flux into the ECS and Redfield ratio (C/N/P = 106:16:1; Redfield, 1958), one can calculate the nutrient-supported marine primary productivity. Since the limiting nutrient for phytoplankton growth was P in the Yangtze input-influenced
- 30 inner shelf and N in the Kuroshio water dominated outer shelf (Chen, 1996; Chen et al., 1996), we use C/P for calculating the Yangtze-sourced, nutrients-supported marine primary productivity, while C/N was used for calculating the Kuroshio-driven nutrients supported marine primary productivity. The results show that primary productivity derived from the Yangtze-sourced nutrients in the ECS is 1.7 × 10⁴² g C yr⁻¹ (DIP: 1.36 × 10⁹ mol yr⁻¹; Chai et al., 2009), while Kuroshio-driven nutrients are responsible for much higher primary productivity, i.e. 16.3 ×

 10^{42} g C yr⁻¹ (DIN: 205 × 10^{9} mol yr⁻¹; Chen, 1996). By using the *in situ* Chl *a* concentration, Gong et al. (2003) estimated the average annual primary production for the ECS continental shelf in 1998 as 145 g C m⁻² yr⁻¹, equal to 72.5 × 10^{42} g C yr⁻¹. This estimation is approximately four times higher than the Yangtze- and Kuroshio water-derived primary productivity achieved in this study, implying the potential role of recycled nutrients from OM remineralization for the primary productivity in the ECS.

If it is the case, then the *in situ* primary productivity in DCM layers should be mainly derived from remineralized nutrients of OM that associated with the surface sediment, as evident from the vertical distribution of turbidity (Fig. 2). This mechanism may result in a higher δ¹³C and C/N in surface sediments than in phytoplankton in DCM

- 10 layers and thus POM due to the preferential remineralization of ⁴²C and N-enriched OM. Consistently, a relatively constant, but higher δ¹³C values and C/N ratios (δ¹³C: -22.4 to -20.1 ‰ and C/N ratio: 6.4-10, unpublished data; Selvaraj Kandasamy) observed in surface sediments collected during the same cruise and at same stations as SPM (Fig. 1), indicating that this mechanism is most likely.
- 15 A step forward, we use the POC concentration (μg L⁻¹), the surface area of the ECS continental shelf with water depths of <200 m (0.5 × 10⁶ km²), and the difference of DCM depths between the inner shelf region (10 m) and the outer shelf region (70 m) (Fig. 2), to estimate the inventory of suspended POC within the DCM layer. Two stations with bottom depths deeper than 200 m (DH7 8: 853 m, DH7 9: 800 m) in the Okinawa Trough were excluded from this estimation. The results show that the maximum (263 μg L⁻¹), minimum (37.3 μg L⁻¹) and mean concentration (92.4 μg L⁻¹) of POC within the DCM interval yields POC inventory of 7.9 × 10¹² g, 1.1 × 10¹² g and

2.8 × 10¹² g, respectively.

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

- 25 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 CO₂, and therefore on phytoplankton δ^{13} C (e.g., Rau et al., 1992; Savoye et al., 2003). The C/N ratio, POC/Chl *a* ratio and δ^{13} C_{POC} all indicated that the POM around the DCM layer is dominated by newly-produced phytoplankton OM (see
- 30 Sections 5.1–5.3). Therefore, to understand the temperature effect on δ¹³C of phytoplankton, we plotted our δ¹³C_{POC} data against temperature into two groups by separating approximately at ~24°C (Fig. 11a). Data points of both groups show a decreasing δ¹³C of phytoplankton biomass while 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 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

- 5 dissolution equilibrium, both may control the concentration of dissolved inorganic carbon in the DCM layers, which are closer to euphotic depths (see Section 4.1). The weak correlation between δ¹³C_{POC} and temperature supports a weak influence of temperature on δ¹³C_{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
- 10 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 -

- 15 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): $\delta^{13}C_{24^{\circ}C} = \delta^{13}C_{POC} - s$ (T – 24), where $\delta^{13}C_{24^{\circ}C}$ 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 $\delta^{13}C_{24^{\circ}C}$ of biomass and POC concentration (circles: R² = 0.71; p<0.0001; n = 18 and triangles: R² = 0.66;
- 20 p<0.0001; n = 18; Fig. 11b), indicating that primary production drives ~70% of the variation of phytoplankton δ¹³C around DCM layers in the southern ECS. On the other hand, δ¹³C_{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.

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<u>5.5</u> Dynamics of δ^{15} N_{PN} in POM in DCM layers

In contrast to <u>the</u> POC and δ¹³C_{POC} relationship (Fig. 10a), there is no significant relationship between PN and its isotopic composition (δ¹⁵N_{PN}) <u>of the POM in SPM</u> investigated <u>in the present study at DCM layers</u> (Fig. 10b), implying that primary productivity has no significant control on <u>the variability of</u> δ¹⁵N_{PN}. As the <u>POM in SPM at around the water depths of</u> DCM layers was dominantly from <u>the</u> newly_produced, phytoplankton-synthesized source, δ¹⁵N_{PN} <u>should be is inferred to be similar to as δ¹⁵N</u> in phytoplankton. Considering the prevalence of low N/P ratio in the DCM layer <u>of in</u> the East China Sea (Lee et al., 2016), the degree of nitrate utilization <u>by for</u> phytoplankton should be high and <u>that</u> would result in the composition of δ¹⁵N_{PN} similar to δ¹⁵N 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 δ¹⁵N_{PN} in DCM layers. Importantly, Tthe spatial distribution of δ^{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.

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According to Li et al. (2010), the range of $\delta^{15}N_{NO3}$ in the Yangtze River was 7.3–12.9 ‰, with a mean value of 8.3 ‰. In the northeast of Taiwan Island, $\delta^{15}N_{NO3}$ 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 $\delta^{15}N_{NO3}$, i.e. higher $\delta^{15}N_{NO3}$ (>6 ‰) in the northeast coastal region and off 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 δ¹⁵N_{PN} variation in the DCM layer of in the East China Sea was primarily governed by the nutrient status and δ¹⁵N_{NO3}⁻, though we do not have nutrient data generated from the same cruise to validate our interpretations.

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There is another possibility that high δ¹⁵N_{PN} (DH7-8: 6.7 ‰, DH7-9: 7.8 ‰) in the DCM layers, off northeast Taiwan (Fig. 5), may not be resulted from the high degree of nitrate utilization, but the incorporation of inorganic nitrogen in <u>the</u>POM. According to Chen et al. (1996) and Liu et al. (1996), NO₃⁻ and NH₄⁺ concentrations in KSSW were high due to the decomposition of OM in sinking particles. However, the concentrations of Chl a Fluorescence as well as POC and PN are low (Figs. 32 and 4). The low Chl a-Fluorescence 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 CO₂ pressure expected to decrease δ^{13} C in DIC that may and also drive a great carbon isotopic fractionation during carbon assimilation by phytoplankton (Rau et al., 1992), a the reason why

δ¹³C_{POC} values in these two stations are low (-25.8 ‰ and -25.2 ‰) compared to values of other locations in the

- study area ECS. Consistently, the low concentration of POC restricts the idea that the high $\delta^{15}N_{PN}$ could not be 25 from the denitrification effect. The high $\delta^{15}N_{PN}$ (6.7 ‰, 7.8 ‰) are probably due to the incorporation of inorganic nitrogen (mainly NH4⁺), the process normally drives can make the δ¹⁵N_{PN} in POM as high as that of inorganic nitrogen δ¹⁵N (Coffin and Cifuentes, 1999). Although δ¹⁵N of NH4⁺ in Kuroshio Water is not available for <u>comparison</u>, according to York et al. (2010), it seems that δ¹⁵N of remineralized NH4⁺ was <u>relatively</u> greater than
- δ^{15} N of NO₃⁻ (York et al., 2010). This possibility is also supported by the high concentrations of NO₃⁻ and NH₄⁺ in 30 Kuroshio Subsurface Water (Liu 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) in of these two SPM samples (DH7-8 and DH7-9).

5.5 Impact of Yangtze River on POM in DCM of ECS

The range of POC/Chl *a* obtained in this study ($33-200 \text{ g g}^{-1}$) 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

- 5 of δ¹³C_{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). Our elemental and isotopic These results of POM indicated that POM around the water depths of DCM was largely derived from the synthesis of *in situ* phytoplankton and the influence of terrestrial OM supplied by the Yangtze River to the in the DCM water depths of southern ECS is lowsignificantly less. The missing of terrestrial OM signals, especially transported by Yangtze River, seems to be related to may be
- 10 because of reservoir and dam buildings along the river in recent years that has shifted believed to shift the location of the Yangtze-derived POC deposition from the inner shelf of the ECS inner shelf to terrestrial reservoirs (Li et al., 2015). The sediment delivered from the river to the estuary has 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 discharged by the Yangtze suspended particles discharge has declined to 150 Mt yr⁻¹,
- 15 less than ~70% of its sediment delivery to the ECS during 1950s. Although 87 % of the mean annual sediment of Yangtze River <u>is was</u>-discharged during the flood season from June to September (Wang et al., 2007; Zhu et al., 2011), <u>among them</u>, approximately 60 <u>out of 87</u>% of the fine-grained sediments are temporarily deposited near the estuary and <u>then</u> later resuspended and transported southward along the inner shelf, off the <u>mainland</u> China <u>mainland coastline</u> (Chen et al., 2017 and references therein). <u>The</u> Yangtze-transported POM moves up toward
- 20 the northeast across the shelf along the so called <u>the a-</u>Changjiang transport pathway in summer season (e.g., Gao et al., 2014), which is <u>largely</u> affected by the combin<u>eding</u> 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. <u>6 and 7</u>) <u>of in this study also <u>illustrate proved</u> this view.</u>
- 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 since 2003, approximately 10% of the world riverine POC burial flux to the oceans (Li et al., 2015). The POC input-flux from the Yangtze into the ECS (range: 1.27–8.5 × 10¹² g C yr⁻¹; Wang et al., 1989; Qi et al., 2014) was significantly less than the estimated primary productivity (72.5 × 10¹² g C yr⁻¹; Gong et al., 2003), implying the predominance of marine-sourced organic matter in the ECSast China Sea. Moreover, the substantial quantityies of organic substances that transported by the Yangtze River may be completely modified before being ultimately deposited onto the inner shelf of the ECSEast China Sea inner shelf 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 index for protein amino acids
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was considered as an indicator for the degree of degradation, ranging from +1 for fresh OM to -1.5 for highly degraded OM (Dauwe and Middelburg, 1998). Wang et al. (2016), based <u>Based</u> on the investigation of lipids biomarkers, in a sediment core collected from the ECS (27.8° N, 122.2° E), <u>Wang et al. (2016)</u> suggested <u>the dominant preservation of that</u> marine autochthonous organic matter dominated the total OM (~90 %) in the ECS.

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Summary and conclusions

In this study, we comprehensively characterized <u>the</u> particulate organic matter (POM) straddling in the depth <u>collected from the deep</u> chlorophyll maximum layers of <u>in the southern</u> East China Sea using hydrographic <u>data</u> 10 parameters (temperature, salinity and turbidity), fluorescence (chlorophyll<u>a</u>) as well as elemental (POC, PN) <u>concentrations</u> and <u>stable</u> carbon and <u>nitrogen</u>-isotopic (δ¹³C_{POC} and δ¹⁵N_{PN}) <u>compositions</u>values. All these parameters indicated that POM <u>around DCM layers wasis</u> dominantly composed of <u>newly-produced OM by</u> <u>phytoplankton with a weak contribution from terrestrial input despite the study area is the best example for the</u>

river-dominated continental margin in the world. We also discussed the main factors controlling the δ^{13} C and δ^{15} N

- 15 variations in phytoplankton in the study area. As for the $\delta^{13}C_{POC}$, 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 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 needs to be substantiated by the nutrient data. Our results show that phytoplankton dynamics drive marine POM composition
- 20 <u>around DCM layers in the southern East China Sea.</u> *in situ* (phytoplankton) newly produced OM with less significant contribution from terrestrial input, although the study area is one of the best examples of riverdominated, shallow continental margins in the world. The δ¹³C in phytoplankton contained relatively low values than that of typical marine phytoplankton (-18 to -20 ‰). Both δ¹³C_{POC} and δ¹⁵N_{PN} show large variations, which are due to the variations in primary productivity and phytoplankton species compositions for δ¹³C_{POC}, and uptake
- 25 of nitrate diffused through the thermocline or locally regenerated ammonia for δ^{45} N_{PN}, respectively.

Moreover, phytoplankton in the southern East China Sea contain relatively low δ¹³C_{POC} values than that of typical marine phytoplankton (–18 to –20 ‰). This study emphasizes the need of sufficient investigation of prerequisite endmember variability and provides a relatively accurate δ¹³C_{POC} and δ¹⁵N_{PN} values of marine endmember in the southern East China Sea, which is crucial for the estimation of relative contributions of terrestrial and marine OM by end_member mixing model. Therefore, our results with highly variable δ¹³C_{POC} and δ¹⁵N_{PN} values in the autotrophic-dominated DCM deep chlorophyll maximum layers can provide an unique ranges for of these two isotopes in the East China Sea, especially the region of 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 during the Holocene in the East China Sea.

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References

30

- Altabet, M. A., and Francois, R.: The use of nitrogen isotopic ratio for reconstruction of past changes in surface ocean nutrient utilization, in: Carbon cycling in the glacial ocean: constraints on the ocean's role in global change, Springer Berlin Heidelberg, 281–306, 1994.
 - Bai, Y., He, X._Q., Chen, C._T._A., Kang, Y., Chen, X., and Caj, W._J.: Summertime Changjiang River plume variation during 1998–2010, J. Geophys. Res., 119, 6238–6257, 2014.
- Beardsley, R. C., Limeburner, R., Yu, H., and Cannon, G. A.: Discharge of the Changjiang (Yangtze River) into the East China Sea, Cont. Shelf Res., 4, 57–76, 1985.
 - Chai, C., Yu, Z., Shen, Z., Song, X. X., Gao, X. H., and Gao, Y.: Nutrient characteristics in the Yangtze River Estuary and the adjacent East China Sea before and after impoundment of the Three Gorges Dam, Sci. Total Environ., 407, 4687–4695, 2009.
- Chang, J., Shiah, F. K., Gong, G. C., and Chiang, K. P.: Cross–shelf variation in carbon-to-chlorophyll a ratios in the East China Sea, summer 1998, Deep-Sea Res. Pt. II, 50, 1237–1247, 2003.
 - Chang, N. N., Shiao, J. C., Gong, G. C., Kao, S. J., and Hsieh, C. H.: Stable isotope ratios reveal food source of benthic fish and crustaceans along a gradient of trophic status in the East China Sea, Cont. Shelf Res., 84, 23– 34, 2014.

- Chen, C. T. A.: Distributions of nutrients in the East China Sea and the South China Sea connection, J. Oceanogr., 64, 737–751, 2008.
- Chen, C. T. A., Lin, C. M., Huang, B. T., and Chang, L. F.: The stoichiometry of carbon, hydrogen, nitrogen, sulfur and oxygen in particular matter of the Western North Pacific marginal seas, Mar. Chem., 54, 179–190, 1996.

Chen, C. T. A.: The Kuroshio intermediate water is the major source of nutrients on the East China Sea continental shelf, Oceanol. Acta, 19, 523–527, 1996.

- Chen, C. T. A., Andreev, A., Kim, K. R., and Yamamoto, M.: Roles of continental shelves and marginal seas in the biogeochemical cycles of the North Pacific Ocean, J. Oceanogr., 60, 17–44, 2004.
- Chen, C. T. A., Kandasamy, S., Chang, Y. P., Bai, Y., He, X. Q., Lu, J. T., and Gao, X. L.: Geochemical evidence of the indirect pathway of terrestrial particulate material transport to the Okinawa Trough, Quat. Int., 441, 51–61, 2017.
- Chen, Y. L. L. and Chen, H. Y.: Nitrate-based new production and its relationship to primary production and chemical hydrography in spring and fall in the East China Sea, Deep-Sea Res. Pt. II, 50, 1249–1264, 2003.

10

Chen, Y. L., Chen, H. Y., Lee, W. H., Hung, C. C., Wong, G. T. F., and Kanda, J.: New production in the East China Sea, comparison between well mixed winter and stratified summer conditions, Cont. Shelf Res., 21, 751–764, 2001.

- Chen, J. and Liu, J.: The spatial and temporal changes of chlorophyll-a and suspended matter in the eastern coastal zones of China during 1997–2013, Cont. Shelf Res., 95, 89–98, 2015.
- Cifuentes, L. A., Sharp, J. H., and Fogel, M. L.: Stable carbon and nitrogen isotope biogeochemistry in the Delaware estuary, Limnol. Oceanogr., 33, 1102–1115, 1988.
- 15 Coffin, R. B. and Cifuentes, L. A.: Stable isotope analysis of carbon cycling in the Perdido Estuary, Florida, Estuaries, 22, 917–926, 1999.
 - Copin-Montegut, C. and Copin-Montegut, G.: Stoichiometry of carbon, nitrogen, and phosphorus in marine particulate matter, Deep-Sea Res. Pt. A, 30, 31–46, 1983.

- 20 particulate carbon to nitrogen ratios in marine surface waters of the Arctic, Global Biogeochem. Cy., 29, 2021– 2033, 2015.
 - Cullen, J. J., Reid, F. M. H., and Stewart, E.: Phytoplankton in the surface and chlorophyll maximum off southern California in August, 1978, J. Plank. Res., 4, 665–694, 1982.

Dai, Z., Du, J., Zhang, X., Su, N., and Li, J.: Variation of Riverine Material Loads and Environmental

- 25 Consequences on the Changjiang (Yangtze) Estuary in Recent Decades (1955--2008), Environ. Sci. Technol., 45, 223-227, 2010.
 - Emerson, S. and Hedges, J. I.: Processes controlling the organic carbon content of open ocean sediments, Paleoceanography, 3, 621–634, 1988.

Fogel, M. L. and Cifuentes, L. A.: Isotope fractionation during primary production, Org. Geochem., 73–98, 1993.

- 30 Fry, B. and Sherr, E. B.: δ¹³C measurements as indicators of carbon flow in marine and freshwater ecosystems, New York: Springer, 196–229, 1989.
 - Fry, B. and Wainright, S. C.: Diatom sources of ¹³C-rich carbon in marine food webs, Mar. Ecol._-Prog. Ser., 149– 157, 1991.

Crawford, D. W., Wyatt, S. N., Wrohan, I. A., Cefarelli, A. O., Giesbrecht, K. E., Kelly, B., and Varela, D. E.: Low

- Furuya, K.: Subsurface chlorophyll maximum in the tropical and subtropical western Pacific Ocean: Vertical profiles of phytoplankton biomass and its relationship with chlorophylla and particulate organic carbon, Mar. Biol., 107, 529–539, 1990.
- Furuya, K., Hayashi, M., Yabushita, Y., and Ishikawa, A.: Phytoplankton dynamics in the East China Sea in
- 5 spring and summer as revealed by HPLC-derived pigment signature, Deep--Sea Res. Pt. II, 50, 367–387, 2003. Gao, L., Li, D. and Ishizaka, J.: Stable isotope ratios of carbon and nitrogen in suspended organic matter: Seasonal and spatial dynamics along the Changjiang (Yangtze River) transport pathway, -J. Geophys. Res., 119, 1717–1737, 2014.
- Gearing, J. N., Gearing, P. J., Rudnick, D. T., Requejo, A. G., and Hutchins, M. J.: Isotopic variability of organic carbon in a phytoplankton-based, temperate estuary, Geochim. Cosmochim. Ac., 48, 1089–1098, 1984.
 - Geider, R. J.: Light and temperature dependence of the carbon to chlorophyll a ratio in microalgae and cyanobacteria: implications for physiology and growth of phytoplankton, New Phytol., 106, 1–34, 1987.
 Geider, R. and La Roche, J.: Redfield revisited: variability of C: N: P in marine microalgae and its biochemical
 - basis, Eur. J. Phycol., 37, 1–17, 2002.

- 15 Gieskes, W. W. C., Kraay, G. W., and Tijssen, S. B.: Chlorophylls and their degradation products in the deep pigment maximum layer of the tropical North Atlantic, Netherlands J. Sea Res., 12, 195–204, 1978.
 - Goericke, R. and Fry, B.: Variations of marine plankton δ¹³C with latitude, temperature, and dissolved CO₂ in the world ocean, Global Biogeochem. Cy., 8, 85–90, 1994.

- Gong, G. C., Wen, Y. H., Wang, B. W., and Liu, G. J.: Seasonal variation of chlorophyll a concentration, primary production and environmental conditions in the subtropical East China Sea, Deep-Sea Res. Pt II, 50, 1219–1236, 2003.
- Gong, G. C., Chang, J., Chiang, K. P., Hsiung, T. M., Hung, C. C., Duan, S. W., and Codispoti, L. A.: Reduction
 of primary production and changing of nutrient ratio in the East China Sea: Effect of the Three Gorges
 Dam?, Geophys. Res. Lett., 33, 2006.
 - Gong, G. C., Shiah, F. K., Liu, K. K., Wen, Y. H., and Liang, M. H.: Spatial and temporal variation of chlorophyll a, primary productivity and chemical hydrography in the southern East China Sea, Cont. Shelf Res., 20, 411–436, 2010.
- 30 Gong, X., Shi, J., Gao, H. W., and Yao, X. H.: Steady-state solutions for subsurface chlorophyll maximum in stratified water columns with a bell-shaped vertical profile of chlorophyll, Biogeosciences, 12, 905–919, 2015.
- Goñi, M. A., Teixeira, M. J., and Perkey, D. W.: Sources and distribution of organic matter in a river-dominated estuary (Winyah Bay, SC, USA), Estuar. Coast. Shelf Sci., 57, 1023–1048, 2003

Gong, G. C. and Liu, G. J.: An empirical primary production model for the East China Sea, Cont. Shelf Res., 23, 213–224, 2003.

- Goñi, M. A., Moore, E., Kurtz, A., Portier, E., Alleau, Y., and Merrell, D.: Organic matter compositions and loadings in soils and sediments along the Fly River, Papua New Guinea, Geochim. Cosmochim. Ac., 140, 275–296, 2014.
- Guo, S. J., Feng, Y. Y., Wang L, Dai, M. H. Liu, Z. L., Bai, Y. and Sun, J.: Seasonal variation in the
 phytoplankton community of a continental-shelf sea: the East China Sea, Mar. Ecol. Prog. Ser., 516, 103–126, 2014a.
 - Guo, C., Liu, H., Zheng, L., Song, S., Chen, B., and Huang, B.: Seasonal and spatial patterns of picophytoplankton growth, grazing and distribution in the East China Sea, Biogeosciences, 11, 1847–1862, 2014b.
- 10 Hale, R. P., Nittrouer, C. A., Liu, J. T., Keil, R. G., and Ogston, A. S.: Effects of a major typhoon on sediment accumulation in Fangliao Submarine Canyon, SW Taiwan, Mar. Geol., 326, 116–130, 2012.

Han, A. Q., Dai, M. H., Gan, J. P., Kao, S. J., Zhao, X. Z., Jan, S., Li, Q., Lin, H., Chen, C. T. A., Wang, L., Hu, J. Y., Wang, L. F., and Gong, F.: Inter-shelf nutrient transport from the East China Sea as a major nutrient source supporting winter primary production on the northeast South China Sea shelf, Biogeosciences, 10, 8159–8170, 2013.

15

20

30

- He, X. Q., Bai, Y., Chen, C. T. A., Hsin, Y. C., Wu, C. R., Zhai, W. D., Liu, Z. L., and Gong, F.: Satellite views of the episodic terrestrial material transport to the southern Okinawa Trough driven by typhoon, J. Geophys. Res., 119, 4490–4504, 2014.
- Head, E. J. H., Harrison, W. G., Irwin, B. I., Horne, E. P. W., and Li, W. K. W.: Plankton dynamics and carbon flux in an area of upwelling off the coast of Morocco, Deep-Sea Res. Pt. I, 43, 1713–1738, 1996.
- Hedges, J. I., Clark, W. A., Quay, P. D., Richey, J. E., Devol, A. H. and Santos, U. D. M.: Compositions and fluxes of particulate organic material in the Amazon River, Limnol. Oceanogr., 31, 717–738, 1986.
- Hedges, J. I., Keil, R. G., and Benner, R.: What happens to terrestrial organic matter in the ocean?, Org. Geochem., 27, 195–212, 1997.
- 25 Hu, D. X. and Yang, Z. S.: Key processes of the ocean flux in the East China Sea, Ocean science publisher, Beijing, 1-204, 2001 (Chinese).
 - Hung, C. C., Tseng, C. W., Gong, G. C. Chen, K. S., Chen, M. H., and Hsu, S. C.: Fluxes of particulate organic carbon in the East China Sea in summer, Biogeosciences, 10, 6469–6484, 2013.

Ichikawa, H. and Beardsley, R. C.: The current system in the Yellow and East China Seas, J. Oceanogr., 58, 77– 92, 2002.

- Isobe, A., and Matsuno, T.: Long-distance nutrient-transport process in the Changjiang river plume on the East China Sea shelf in summer, J. Geophys. Res., 113, C04006, doi:10.1029/2007JC004248, 2008.
- Isobe, A., Fujiwara, Y., Chang, P. H., Sugimatsu, K., Shimizu, M., Matsuno, T., and Manda, A.: Intrusion of less saline shelf water into the Kuroshio subsurface layer in the East China Sea, J. Oceanogr., 60, 853–863, 2004.

- Jiang, Z., Chen, J., Zhou, F., Shou, L., Chen, Q., Tao, B., Yan, X., and Wang, K.: Controlling factors of summer phytoplankton community in the Changjiang (Yangtze River) Estuary and adjacent East China Sea shelf, Cont. Shelf Res., 101, 71–84, 2015.
- Jiang, Z., Chen, J., Zhou, F., Zhai, H., Zhang, D., and Yan, X.: Summer distribution patterns of Trichodesmium spp. in the Changjiang (Yangtze River) Estuary and adjacent East China Sea shelf, Oceanologia, <u>59, 248–261,</u> 2017.
- Kao, S. J., Lin, F. J., and Liu, K. K.: Organic carbon and nitrogen contents and their isotopic compositions in surficial sediments from the East China Sea shelf and the southern Okinawa Trough, Deep-Sea Res. Pt. II, 50, 1203–1217, 2003.
- 10 Karlson, B., Edler, L., Granéli, W., Sahlsten, E., and Kuylenstierna, M.: Subsurface chlorophyll maxima in the Skagerrak-processes and plankton community structure, J. Sea Res., 35, 139–158, 1996.
 - Katoh, O., Morinaga, K., and Nakagawa, N.: Current distributions in the southern East China Sea in summer, J. Geophys. Res., 105, 8565–8573, 2000.

Kirk, J. T.: Light and photosynthesis in aquatic ecosystems, Cambridge university press, 1994.

5

15 Kopczyńska, E. E., Goeyens, L., Semeneh, M., and Dehairs, F.: Phytoplankton composition and cell carbon distribution in Prydz Bay, Antarctica: relation to organic particulate matter and its δ¹³C values, J. Plankton Res., 17, 685–707, 1995.

- 20 Lee, K. J., Matsuno, T., Endoh, T., Endoh, T., Ishizaka, J., Zhu, Y. L., Takeda, S. and Sukigara C.: A role of vertical mixing on nutrient supply into the subsurface chlorophyll maximum in the shelf region of the East China Sea, Cont. Shelf Res., 2016.
 - Letelier, R. and Karl, D._M.: Role of Trichodesmium spp. in the productivity of the subtropical North Pacific Ocean, Mar. Ecol. Prog. Ser., 133, 263–273, 1996.
- 25 Letelier, R. and Karl, D._M.: Trichodesmium spp. physiology and nutrient fluxes in the North Pacific subtropical gyre, Aquat. Microbial Ecol., 15, 265–276, 1998.
 - Li, G., Wang, X. T., Yang, Z., Mao, C., West, A. J., and Ji, J.: Dam-triggered organic carbon sequestration makes the Changjiang (Yangtze) river basin (China) a significant carbon sink, J. Geophys. Res, 120, 39–53, 2015.

Li, S. L., Liu, C. Q., Li, J., Liu, X., Chetelat, B., Wang, B., and Wang, F.: Assessment of the sources of nitrate in

- 30 the Changjiang River, China using a nitrogen and oxygen isotopic approach, Environ. Sci. Technol., 44, 1573– 1578, 2010.
 - Li, Y. H.: Material exchange between the East China Sea and the Kuroshio current, Terr. Atmos. Oceanic Sci., 5, 625–631, 1994.

Lee, H. J. and Chao, S. Y. A climatological description of circulation in and around the East China Sea, Deep-Sea Res. Pt. II, 50, 1065–1084, 2003.

Lie, H. J., Cho, C. H., Lee, J. H., and Lee, S.: Structure and seaward extension of the Changjiang River plume in the East China Sea, J. Geophys. Res., 108, 3077, 2003.

Liénart, C., Susperregui, N., Rouaud, V., Cavalheiro, J., David, V., Del Amo, Y., Duran, R., Lauga, B., Monperrus, M., Pigot, T., Bichon, S., Charlier, K., and Savoye, N.: Dynamics of particulate organic matter in a coastal system characterized by the occurrence of marine mucilage-A stable isotope study, J. Sea Res., 116, 12–22,

- system characterized by the occurrence of marine mucilage-A stable isotope study, J. Sea Res., 116, 12–22, 2016.
 - Liu, J. P., Li, A. C., Xu, K. H., Velozzi, D. M., Yang, Z. S., Milliman, J. D., and DeMaster, D. J.: Sedimentary features of the Yangtze River-derived along-shelf clinoform deposit in the East China Sea, Cont. Shelf Res., 26, 2141–2156, 2006.
- 10 Liu, J. P., Xu, K. H., Li, A. C., Milliman, J. D., Velozzi, D. M., Xiao, S. B. and Yang, Z. S.: Flux and fate of Yangtze River sediment delivered to the East China Sea, Geomorphology, 85, 208–224, 2007.
 - Liu, K. K., Pai, S. C., and Liu, C. T.: Temperature-nutrient relationships in the Kuroshio and adjacent waters near Taiwan, Acta Oceanogr. Taiwan., 21, I–17, 1988.

Liu, K. K., Su, M. J., Hsueh, C. R., and Gong, G. C.: The nitrogen isotopic composition of nitrate in the Kuroshio

- 15 Water northeast of Taiwan: Evidence for nitrogen fixation as a source of isotopically light nitrate, Mar. Chem., 54, 273–292, 1996.
 - Liu, W. C., Wang, R., and Li, C. L.: C/N ratios of particulate organic matter in the East China Sea, Oceanologia et Limnologia Sinica, 29, 467–470, 1998 (Chinese).

Lorenzen, C. J.: Vertical distribution of chlorophyll and phaeo-pigments: Baja California, In Deep Sea Research and Oceanographic Abstracts, Elsevier, 14, 735–745, 1967.

Lorrain, A., Savoye, N., Chauvaud, L., Paulet, Y. –M., and Naulet, N.: Decarbonation and preservation method for the analysis of organic C and N contents and stable isotope ratios of low-carbonated suspended particulate material, Anal. Chim. Acta, 491, 125–133, 2003.

20

Lowe, A. T., Galloway, A. W. E., Yeung, J. S., Dethier, M. N., and Duggins, D. O.: Broad sampling and diverse
 biomarkers allow characterization of nearshore particulate organic matter, Oikos, 123, 1341–1354, 2014.

Mague, T. H., Mague, F. C., and Holm-Hansen, O.: Physiology and chemical composition of nitrogen fixing phytoplankton in the central North Pacific Ocean, Mar. Biol., 41, 75–82, 1977.

Malone, T. C., Falkowski, P. G., Hopkins, T. S., Rowe, G. T., and Whitledge, T. E.: Mesoscale response of diatom populations to a wind event in the plume of the Hudson River, Deep-Sea Res., 30, 149–170, 1983.

- 30 Martiny, A. C., Pham, C. T., Primeau, F. W., Vrugt, J. A., Moore, J. K., Levin, S. A., and Lomas, M. W.: Strong latitudinal patterns in the elemental ratios of marine plankton and organic matter, Nat. Geosci., 6, 279–283, doi:10.1038/ngeo1757, 2013a.
 - Meyers, P. A.: Preservation of elemental and isotopic source identification of sedimentary organic matter, Chem. Geol., 114, 289–302, 1994.

<u>Miller, R. J., Page, H. M., and Brzezinski, M. A.: δ¹³C and δ¹⁵N of particulate organic matter in the Santa Barbara</u> <u>Channel: drivers and implications for trophic inference, Mar. Ecol. Prog. Ser., 474, 53–66, 2013.</u>

Milliman, J. D. and Farnsworth. K. L.: River Discharge to the Coastal Ocean: A Global Synthesis, Cambridge Univ. Press, 2011.

- 5 Minagawa, M., Ohashi, M., Kuramoto, T., and Noda, N.: δ¹⁵N of PON and nitrate as a clue to the origin and transformation of nitrogen in the subarctic North Pacific and its marginal sea, J. Oceanogr., 57, 285–300, 2001.
 Montagnes, D. J., Berges, J. A., Harrison, P. J. and Taylor, F.: Estimating carbon, nitrogen, protein, and chlorophyll a from volume in marine phytoplankton, Limnol. Oceanogr., 39, 1044–1060, 1994.
- Nakatsuka, T., Handa, N., Wada, E., and Wong, C. S.: The dynamic changes of stable isotopic ratios of carbon
- 10 and nitrogen in suspended and sedimented particulate organic matter during a phytoplankton bloom, J. Mar. Res., 50, 267–296, 1992.
 - Nielsen, E. S. and Hansen, V. K.: Light adaptation in marine phytoplankton populations and its interrelation with temperature, Physiol. Plant., 12, 353–370, 1959.
- Peters, K. E., Sweeney, R. E., and Kaplan, I. R.: Correlation of carbon and nitrogen stable isotope ratios in sedimentary organic matter, Limnol. Oceanogr., 23, 598–604, 1978.
- Pilati, A. and Wurtsbaugh, W. A.: Importance of zooplankton for the persistence of a deep chlorophyll layer: a limnocorral experiment, Limnol. Oceanogr., 48, 249–260, 2003.
 - Qi, W., Müller, B., Pernet-Coudrier, B., Singer, H., Liu, H., Qu, J., and Berg, M.: Organic micropollutants in the Yangtze River: seasonal occurrence and annual loads, Sci. Total Environ., 472, 789–799, 2014.
- 20 Rau, G. H., Takahashi, T., Des Marais, D. J., Repeta, D. J., and Martin, J. H.: The relationship between δ¹³C of organic matter and [CO₂ (aq)] in ocean surface water: data from a JGOFS site in the northeast Atlantic Ocean and a model, Geochim. Cosmochim. Ac., 56, 1413–1419, 1992.
- Ravichandran, M., Girishkumar, M. S., and Riser, S.: Observed variability of chlorophyll-a using Argo profiling floats in the southeastern Arabian Sea, Deep-Sea Res. Pt. I, 65, 15–25, 2012.
- Redfield, A. C.: The biological control of chemical factors in the environment, Am. Sci., 46, 230A205–221, 1958.
 Riley, G. A., Stommel, H. M., and Bumpus, D. F.: Quantitative ecology of the plankton of the western North Atlantic, Bull. Bingham Oceanogr. Coll., 12, 1–169, 1949.

Ryabov, A. B., Rudolf, L., and Blasius, B.: Vertical distribution and composition of phytoplankton under the influence of an upper mixed layer, J. Theor. Biol., 263, 120–133, 2010.

30 Savoye, N., Aminot, A., Treguer, P., Fontugne, M., Naulet, N., and Kerouel, R.: Dynamics of particulate organic matter δ¹⁵N and δ¹³C during spring phytoplankton blooms in a macrotidal ecosystem (Bay of Seine, France)₋ Mar. Ecol.-Prog. Ser., 255, 27–41, 2003.

- Savoye, N., David, V., Morisseau, F., Etcheber, H., Abril, G., Billy, I., Charlier, K., Oggian, G., Derriennic, H., and Sautour, B.: Origin and composition of particulate organicmatter in a macrotidal turbid estuary: The Gironde estuary France, Estuar. Coast. Shelf Sci., 108, 16–28, 2012.
- Selvaraj, K., Lee, T. Y., Yang, J. Y. T., Canuel, E. A., Huang, J. C., Dai, M., Liu, J. T., and Kao, S. J.: Stable isotopic and biomarker evidence of terrigenous organic matter export to the deep sea during tropical storms, Mar. Geol., 364, 32–42, 2015.
- Sharples, J., Moore, C. M., Rippeth, T. P., Holligan, P. M., Hydes, D. J., Fisher, N. R., and Simpson, J. H.: Phytoplankton distribution and survival in the thermocline, Limnol. Oceanogr., 46, 486–496, 2001.
- Sheu, D. D., Jou, W. C., Chung, Y. C., Tang, T. Y. and Hung, J. J.: Geochemical and carbon isotopic characterization of particles collected in sediment traps from the East China Sea continental slope and the Okinawa Trough northeast of Taiwan, Cont. Shelf Res., 19, 183–203, 1999.
 - Sigman, D. M., Karsh, K. L., and Casciotti, K. L.: Ocean process tracers: nitrogen isotopes in the ocean, in: Encyclopedia of ocean science, 2nd ed., edited by Steele, J. H., Turekian, K. K., Thorpe, S. A., Academic Press, London, 40–54, 2009
- Su, J. L., and Y. Q. Pan, Y. Q.: On the shelf circulation north of Taiwan, Acta Oceanol. Sin., 6, 1–20, 1987.
 Tan, S. C., Shi, G. Y., Shi, J. H., Gao, H. W., and Yao, X.: Correlation of Asian dust with chlorophyll and primary productivity in the coastal seas of China during the period from 1998 to 2008, J. Geophys. Res, 116, 2011.
 - Tang, S., Chen, C., Zhan, H., <u>and Xu</u>, D.: Remotely-sensed estimation of the euphotic depth in the northern South China Sea, Geoscience and Remote Sensing Symposium, 2007. IGARSS 2007. IEEE International, 23–
- 20 28 July 2007, Barcelona, http://dx.doi.org/10.1109/IGARSS.2007.4422947, 2007.

- Thompson, P. A., Guo, M. X. and Harrison, P. J.: Effects of variation in temperature. I. On the biochemical composition of eight species of marine phytoplankton, J. Phycol., 28, 481–488, 1992.
- Thornton, S. F. and McManus, J.: Application of organic carbon and nitrogen stable isotope and C/N ratios as source indicators of organic matter provenance in estuarine systems: evidence from the Tay Estuary,
 Scotland, Estuar. Coast. Shelf Sci., 38, 219–233, 1994.
- Umezawa, Y., Yamaguchi, A., Ishizaka, J., Hasegawa, T., Yoshimizu, C., Tayasu, I., Yoshimura H., Morii, I., Aoshima, T., and Yamawaki, N.: Seasonal shifts in the contributions of the Changjiang River and the Kuroshio Current to nitrate dynamics in the continental shelf of the northern East China Sea based on a nitrate dual isotopic composition approach, Biogeosciences, 11, 1297–1317, 2014.
- 30 Wada, E., Minagawa, M., Mizutani, H., Tsuji, T., Imaizumi, R., and Karasawa, K.: Biogeochemical studies on the transport of organic matter along the Otsuchi River watershed, Japan, Estuar. Coast. Shelf Sci., 25, 321–336, 1987.

- Wang M. Y., Zhao, G. J. and Zhang, S. The transport of carbon, nitrogen, phosphorus and sulfur in the Changjiang, The Background Study of Chemical Elements in the Water Environments, Mapping Publisher, Beijing, 122–131, 1989.
- Wang, R., Wang, J., Li, F., Yang, S., and Tan, L.: Vertical distribution and indications of lipids biomarkers in the sediment core from East China Sea, Cont. Shelf Res., 122, 43–50, 2016.

- Wang, Z. H., Li, L. Q., Chen, D. C., Xu, K. Q., Wei, T. Y., Gao, J. H., Zhao, Y. W., Chen, Z. Y., and Masabate, W.: Plume front and suspended sediment dispersal off the Yangtze (Changjiang) River mouth, China during nonflood season, Estuar. Coast. Shelf Sci., 71, 60–67, 2007.
- Weston, K., Fernand, L., Mills, D. K., Delahunty, R., and Brown, J.: Primary production in the deep chlorophyll
 maximum of the central North Sea, J. Plankton Res., 27, 909–922, 2005.
 - Wong, G. T. F., Pai, S. C., Liu K. K., Liu, C. T., and Chen, C. T. A.: Variability of the chemical hydrography at the frontal region between the East China Sea and the Kuroshio north-east of Taiwan, Estuar. Coast. Shelf Sci., 33, 105–120, 1991.
- Wong, G. T. F., Chao, S. Y., Li, Y. H., and Shiah, F. K.: The Kuroshio edge exchange processes (KEEP) studyan introduction to hypotheses and highlights, Cont. Shelf Res., 20, 335–347, 2000.
 - Wu, Y., Zhang, J., Li, D. J., Wei, H., and Lu, R. X.: Isotope variability of particulate organic matter at the PN section in the East China Sea, Biogeochemistry, 65, 31–49, 2003.
 - Wu, Y., Dittmar, T., Ludwichowski, K. U., Kattner, G., Zhang, J., Zhu, Z. Y., and Koch, B. P.: Tracing suspended organic nitrogen from the Yangtze River catchment into the East China Sea, Mar. Chem., 107, 367–377, 2007a.
- 20 Wu, Y., Zhang, J., Liu, S. M., Zhang, Z. F., Yao, Q. Z., Hong, G. H., and Cooper, L.: Sources and distribution of carbon within the Yangtze River system, Estuar., Coast. Shelf Sci., 71, 13–25, 2007b.
 - Wu, Y., Eglinton, T., Yang, L., Deng, B., Montluçon, D., and Zhang, J.: Spatial variability in the abundance, composition, and age of organic matter in surficial sediments of the East China Sea, J. Geophys. Res.,118, 1495–1507, 2013.
- 25 Yang, S. L., J. Zhang, J., Zhu, J., Smith, J. P., Dai, S. B., Gao, A., and Li, P.: Impact of dams on Yangtze River sediment supply to the sea and delta intertidal wetland response, J. Geophys. Res., 110, F03006, doi:10.1029/2004JF000271, 2005.
 - Yang, S. L., Milliman, J. D., Li, P., and Xu, K.: 50,000 dams later: erosion of the Yangtze River and its delta₋₋ Global Planet. Change, 75, 14-20, 2011.
- 30 York, J. K., Tomasky, G., Valiela, I., and Giblin, A. E.: Isotopic approach to determining the fate of ammonium regenerated from sediments in a eutrophic sub-estuary of Waquoit Bay, MA., Estuar. Coast., 33, 1069–1079, 2010.

- Zhang, J., Wu, Y., Jennerjahn, T. C., Ittekkot, V., and He, Q.: Distribution of organic matter in the Changjiang (Yangtze River) Estuary and their stable carbon and nitrogen isotopic ratios: Implications for source discrimination and sedimentary dynamics, Mar. Chem., 106, 111–126, 2007.
- Zheng, L., Chen, B., Liu, X., Huang, B., Liu, H., and Song, S.: Seasonal variations in the effect of microzooplankton grazing on phytoplankton in the East China Sea, Cont. Shelf Res., 111, 304–315, 2015.

10

- Zhou, F., Xue, H., Huang, D., Xuan, J., Ni, X., Xiu, P., and Hao, Q.: Cross-shelf exchange in the shelf of the East China Sea, J. Geophys. Res., 120, 1545–1572, 2015.
- Zhou, W. H., Yin, K. D., Long, A. M., Huang, H., Huang, L. M., and Zhu, D. D.: Spatial-temporal variability of total and size-fractionated phytoplankton biomass in the Yangtze River Estuary and adjacent East China Sea coastal waters, China, Aquat. Ecosyst. Health Manage., 15, 200–209, 2012.
- Zhu, C., Wang, Z. H., Xue, B., Yu, P. S., Pan, J. M., Wagner, T., and Pancost, R. D.: Characterizing the depositional settings for sedimentary organic matter distributions in the Lower Yangtze River-East China Sea Shelf System, Estuar. Coast. Shelf Sci., 93, 182–191, 2011.

Table 1. Summary statistics of elemental and isotopic compositions of suspended particulate matter (SPM) at around DCM layers in the continental shelf region of East China Sea (n=36).

	Sampling								
	depth	SPM	δ ¹³ C _{POC}	δ ¹⁵ ₩₽Ν	POC	PN	POC	PN	C/N
	m	mg L ^{_1}	%₀	<u>0/</u>	<mark>µg L</mark> -¹	<mark>µg L</mark> −¹	%	<u>%</u>	molar
Min	10	1.7	-25.8	3.8	20.4	4.5	0.90	0.19	4.1
Max	130	14.7		7.8	263.0	52.8	4.74	0.95	6.3
Mean	45	4.4	-23.1	6.1	85.5	17.8	2.17	0.45	5.6
SD	21	2.7	1.45	1.0	49.5	9.94	0.94	0.18	0.5

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 *a* Fluorescence. SD=Standard deviation.

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

Table 2. POC/Chl a in the Western Pacific compiled from the liter	ature.

Region	Depth (m)	POC/Chl a (g g ⁻¹)	Method	Note	References
ECS	DCM	35.3	POC vs. Chl a regression	R² = 0.49, p<0.0001	This study
ECS	2—150	64	POC vs. Chl a regression	R² = 0.61, p<0.001	Hung et al., 2013
Coastal of ECS	2/5	13	POC vs. Chl a regression	R² = 0.43, p<0.05	Chang et al., 2003
Middle shelf and Kuroshio zone of ECS	2	93	POC vs. Chl a regression	R² = 0.65, p<0.001	Chang et al., 2003
Coastal zone of ECS	5	18	Cell volume		Chang et al., 2003
Middle shelf of ECS	5	67	Cell volume		Chang et al., 2003
Kuroshio zone of ECS	5	9 4	Cell volume		Chang et al., 2003
Western Pacific	ÐCM	52	Cell volume		Furuya, 1990

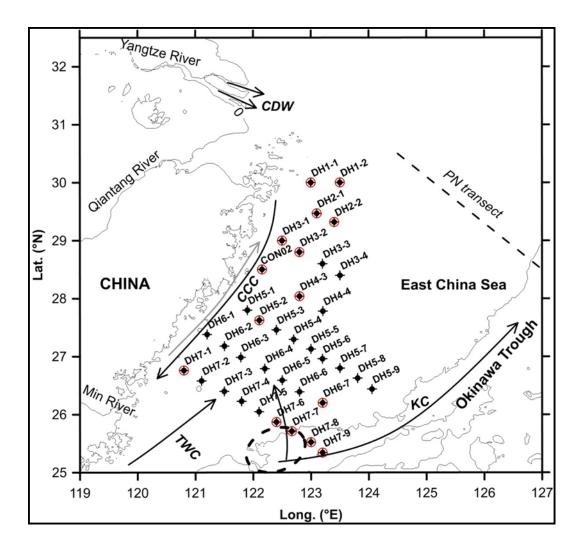


Figure 1. Map showing the locations of suspended particulate matters (SPM) collected <u>around</u> the deep chlorophyll maximum (DCM) layer from the southern East China Sea during in summer (June 22–July 21) 2013 for the present investigation. Also shown is the along with modern current patterns in the East China Sea. Also shown are station names along with locations. Red circles mark the SPM samples that were collected from the water depths either below or above but mostly contiguous to the DCM layer. CDW – Changjiang Diluted Water, CCC – China Coast Current, TWC – Taiwan Warm Current and KC – Kuroshio Current. The dashed ellipse represents the center of Kuroshio upwelling, <u>occurring</u> due to an abrupt change in the bottom topography, in the northeast of Taiwan Island (Wong et al., 2000). Also shown is the *PN* transect, a cross shelf transect that is relatively well studied for particulate organic matter dynamics in the East China Sea.

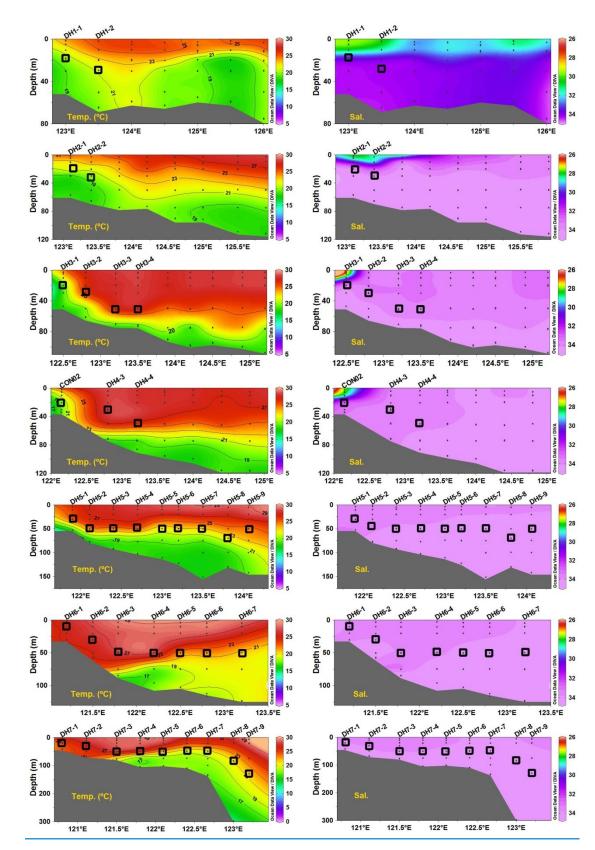
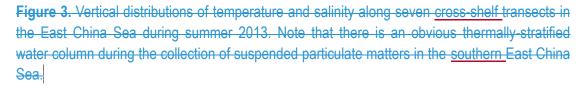


Figure 2. Vertical distributions of temperature and salinity along seven transects across the southern East China Sea during summer 2013. Note that there is an obvious thermally-stratified water column during the collection of suspended particles in the study area.



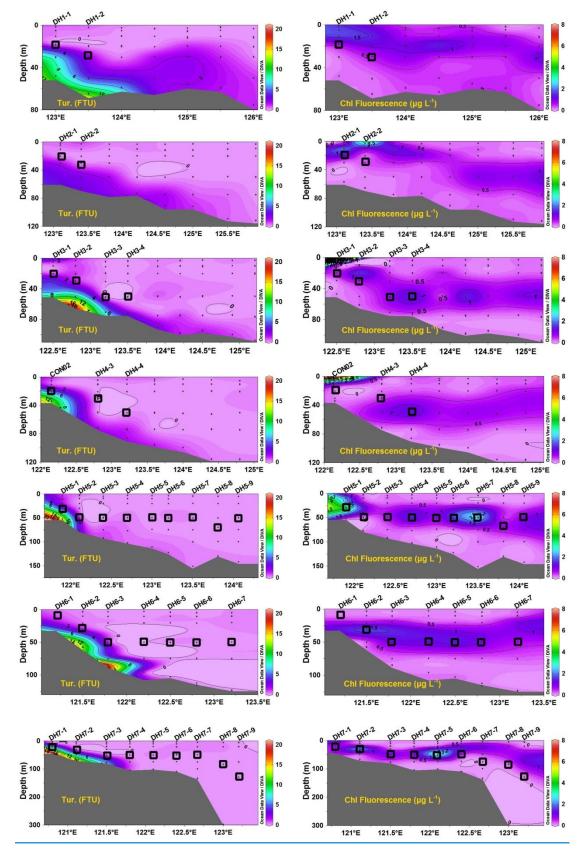


Figure 3. Vertical distributions of turbidity (Tur.) and chlorophyll fluorescence (Chl Fluorescence) concentration along seven cross-shelf transects in the southern East China Sea during summer 2013.

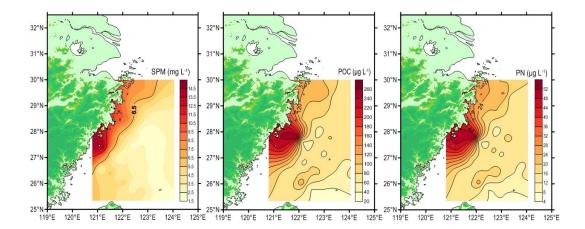


Figure 4. Spatial distributions of suspended particulate matters (SPM, mg L⁻¹), particulate organic carbon (POC, μ g L⁻¹) and <u>particulate</u> nitrogen (PN, μ g L⁻¹) at around the deep chlorophyll maximum layer in the <u>southern</u> East China Sea <u>during in</u>-summer 2013.

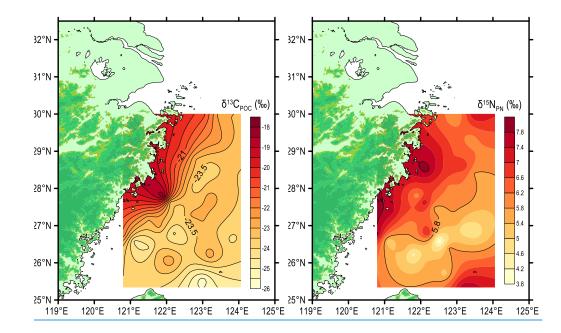


Figure 5. Spatial distributions of stable isotopic values of particulate organic carbon and nitrogen ($\delta^{13}C_{POC}$ and $\delta^{15}N_{PN}$) in suspended particulate matters at around the deep chlorophyll maximum layers in the southern East China Sea <u>during in</u> summer 2013.

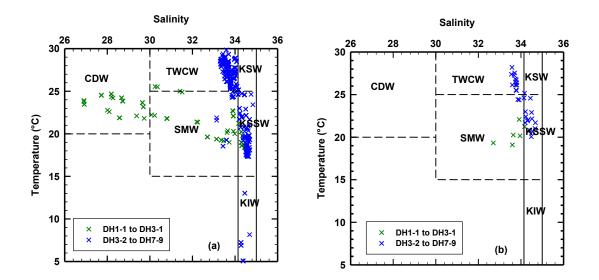


Figure 6. Temperature–Salinity (*T–S*) diagrams for (a) the entire water column in the East China Sea and (b) the deep chlorophyll maximum layers where from the suspended particulate matters were collected for the present investigation. *T–S* ranges of seven-six water masses are taken from Umezawa *et al.* (2014). <u>CDW – Changjiang Diluted Water;</u> TWCW – Taiwan Warm Current Water; SMW – Shelf Mixed Water; KSW – Kuroshio Surface Water; KSSW – Kuroshio Subsurface Water; KIW – Kuroshio Intermediate Water.

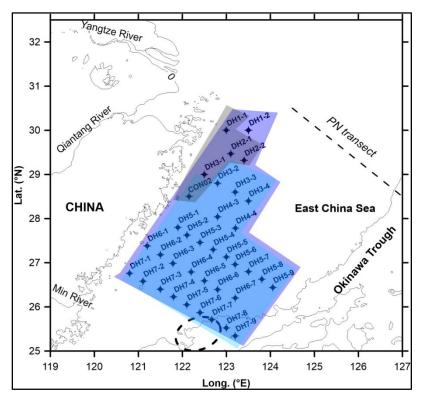


Figure 7. <u>A</u><u>D</u><u>d</u>iagram delineating <u>the</u> regions influenced by three main water masses based on the *T*-<u>S</u> <u>relationship</u> (Figs. <u>3-2</u> and 6) in the study area. Area with <u>greyblack</u> polygon represents the <u>influence of CDW-influence</u>, which is limited only in the upper 10 m₂₇ <u>aA</u>rea with sky blue represents the <u>dominance of TWCW-dominance</u>, which <u>is limited to influenced</u>~30 m below the surface<u>.</u>, and t<u>T</u>he polygon colored by deep blue represents the area influenced by <u>the</u> KSSW, <u>indicating which influenced that</u> the bottom water of the entire study area<u>was</u> <u>dominated by KSSW</u>.

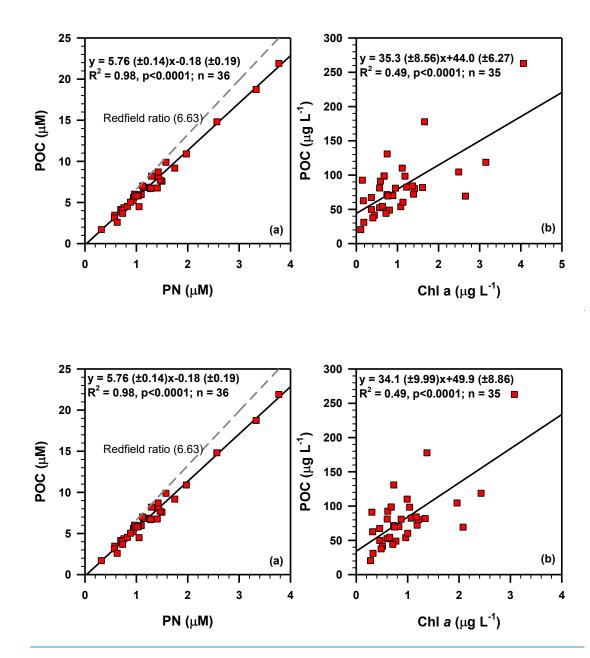


Figure 8. Bi-plots showing the relationships of (a) <u>POC vs.</u> PN <u>vs. POC</u> and (b) <u>POC vs.</u> Chl *a* <u>vs. POC</u> in suspended particulate matters investigated in this study. Redfield ratio (dashed line in panel a) is taken from Redfile!d (1958).

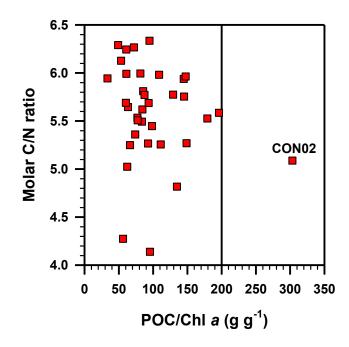


Figure 9. <u>Molar C/N ratio vs.</u> POC/Chl *a* <u>vs. molar C/N</u>-ratio in suspended particulates investigated in this study. The vertical line represents POC/Chl *a* ratio of 200 g g⁻¹, which is the upper limit of this ratio for phytoplankton-dominated particulate organic matter (Savoye et al., 2003). See text for more details. CON02 is the station where red tide was observed during the sampling time and the color of the surface water was brown and dissolved oxygen in the bottom water was 1.6 mg L⁻¹.

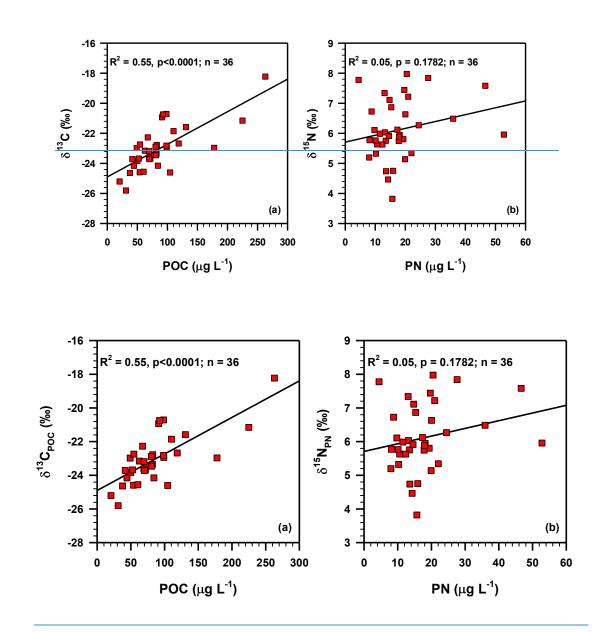


Figure 10. Bi-plots showing the relationships of (a) $\underline{\delta^{13}C_{POC}}$ vs. POC vs. $\overline{\delta^{13}C_{POC}}$ and (b) $\underline{\delta^{15}N_{PN}}$ vs. PN vs. PN vs. $\overline{\delta^{15}N_{PN}}$ in suspended particulate matters <u>around</u> from the deep chlorophyll maximum layers in the <u>southern</u> East China Sea.

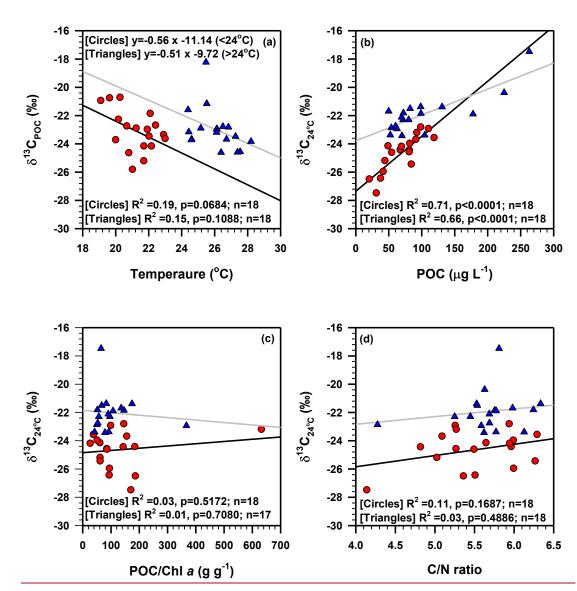


Figure 11. Bi-plots showing the relationships of (a) $\delta^{13}C_{POC}$ vs. temperature for samples separated into two based on temperature: <24°C and >24°C, (b) temperature-normalized $\delta^{13}C_{24^{\circ}C}$ vs. POC concentration, (c) $\delta^{13}C_{24^{\circ}C}$ vs. POC/Chl *a* ratio and (d) $\delta^{13}C_{24^{\circ}C}$ vs. molar C/N ratio in suspended particulate matters around deep chlorophyll maximum layers in the southern East China Sea.