

General overview:

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

My primary concerns with the manuscript are as following:

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

Specific points:

Introduction

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

“Stable isotopes of organic carbon and nitrogen ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) and molar carbon to nitrogen (C/N) ratios are natural tracers frequently used to identify the source and fate of terrestrial organic matter (OM) in estuarine and marine environments (Meyers, 1994; Hedges et al., 1997; Goñi et al., 2014; Selvaraj et al., 2015). This approach is based on $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and C/N ratios being significantly different between different end-members (e.g. terrestrial and marine), and the assumption that only conservative physical mixing of bulk properties occur in these marginal settings (Thornton and McManus, 1994; Hedges et al., 1986). Quantifying the relative contribution of end-members using mass balance models thus requires known and constant elemental and isotopic values of end-members and major sources of OM in the study region (e.g., Goñi et al., 2003). Therefore, application of mixing models for the discrimination of OM sources discrimination requires clearly identified representative values for local OM sources. However, often end-member values of $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and molar C/N ratios are represented by ‘typical’ numbers, such as ca. -20‰ and -27‰ for $\delta^{13}\text{C}$ of marine phytoplankton and terrestrial plants [INSERT SUPPORTING REFERENCE], respectively, but without measuring discrete end-member values in real, local or regional OM source materials. For example, a number of earlier studies failed to measure isotopic values of marine phytoplankton despite using end-member mixing models to distinguish marine versus terrestrial OM in surface sediments (e.g., Kao et al., 2003; Wu et al., 2013), or distinguish marine phytoplankton values from

bulk surface particulate organic matter (POM) values (e.g., Zhang et al., 2007), or allochthonous POM (e.g., Hale et al., 2012). As POM in estuarine and marine systems is mostly derived from primary production, the stable isotope values ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) and molar C/N ratios of POM are largely representative of phytoplankton biomass (Gearing et al., 1984). Therefore, since phytoplankton are the main primary producers of marine OM, the elemental and isotopic compositions of phytoplankton should be considered while studying the dynamics of POM in the marine water column.”

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

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

Sharples et al. (2001) Phytoplankton distribution and survival in the thermocline, L&O, DOI: 10.4319/lo.2001.46.3.0486

Moore et al. (2006) Phytoplankton photoacclimation and photoadaptation in response to environmental gradients in a shelf sea, L&O DOI: 10.4319/lo.2006.51.2.0936

Hickman et al. (2012) Primary production and nitrate uptake within the seasonal thermocline of a stratified shelf sea, MEPS, DOI: 10.3354/meps09836

Williams et al. (2013) Wind-driven nutrient pulses to the subsurface chlorophyll maximum in seasonally stratified shelf seas, GRL, DOI: 10.1002/2013GL058171

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

P3, L14: Suggest the following grammatical changes: “Nutrient-rich freshwater inputs in turn stimulate water column productivity in coastal water compared to the open ocean. Annual primary production over the entire shelf of the East China Sea is high relative to other marginal seas and was estimated to be”

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

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

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

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

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

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

P3, L29: Replace “comprehend” with “determine”.

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

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

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

P4, L12: Replace “middle” with “central”. Is “north” intentionally repeated?

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

P4, L16: Replace “it has been believed” with “it is thought that”, perhaps replace “source” with “component”.

P4, L16: Suggesting the following: “In winter, the CDW flows southwards along the coastline of a mainland China as a narrow jet (Chen, 2008; Han et al., 2013) and in summer spreads to the northeast (Isobe et al., 2004). These changes are driven by the East Asian monsoon, which constitutes a strong northeast monsoon in winter and weaker southwest monsoon in summer.”

P4, L19: Insert “The” before “Taiwan Warm Current”.

P4, L20: Delete “the” before “intruding”.

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

P4, L24: Perhaps “oxygen-under saturated”. Delete “but”.

P4, L25: Replace “East China Sea” with “ECS”.

P4, L27: Suggest: “Furthermore, Kuroshio water accounts for up to 90 % of shelf waters in the ECS...”

P4, L30: Suggest: “Primary production in the ECS is nutrient-limited in summer and light-limited in winter (Chen et al., 2001; Chen and Chen 2003), with production being higher in summer. In 2008, annual primary production rates showed distinct spatial variation, with rates in the north-western ECS ($155 \text{ g C m}^{-2} \text{ y}^{-1}$) being higher than those of the south-eastern ECS ($144 \text{ g C m}^{-2} \text{ y}^{-1}$) and the overall average for the ECS ($145 \text{ g C m}^{-2} \text{ y}^{-1}$) (Gong et al., 2003). However, primary production rates have decreased by 86 % between 2008 and 2003, due to installation of a number of reservoirs within the Yangtze River drainage basin (Gong et al., 2006).

Material and methods

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

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

P5, L12 - 20: More detail is needed with respect to sample collection. How many depths did you sample during each cast? What auxiliary samples were collected, e.g. Chl a, dissolved inorganic nutrients etc.? What volume were the Niskin bottles? How were samples collected from the CTD – did you use any tubing and if so what kind? How were the PVC bottles cleaned prior to receiving samples? What volume were the PVC bottles? Approximately how long were samples stored prior to filtering? What vacuum pressure/positive pressure conditions did you filter under? What was the range of volumes that you filtered for each sample, e.g. was it of the order of 1L, 2L, 10L? If you did not rinse the filters prior to storage do you think that the potential contribution of salts to the SPM weights was negligible for the volumes you were filtering? Details like these will give the reader more confidence in your results.

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

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

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

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

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

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

P6, L8: Replace “is” with “was”.

P6, L14 – 23: You have raised this issue that your results may show some bias due to de-carbonation of the PN filters and you have provided evidence that your results are in line with those of Wu et al. (2003), who also de-carbonated their samples from this study region. It would perhaps be more useful to be able to quote similar values for this region obtained without de-carbonation of PN filters to suggest that the bias resulting from the freezing and de-carbonation process did not significantly alter your results.

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

Results and interpretations

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

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

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

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

P7, L19: Replace “limited along the coast” with “restricted to coastal stations”.

P7, L27-28: Suggest restructuring to “The highest Chl fluorescence concentration ($18.0 \mu\text{g L}^{-1}$) was observed in surface waters at station DH3-1. All other values were less than $8.0 \mu\text{g L}^{-1}$ (Fig. 3).”

P7, L28: “were” not “are”

P7, L29: “showed” not “show”

P7, L32: Replace “straddling around” with “across”

P7, L33: Delete the comma after depth

P8, L7: Delete “productivity”.

P8, L10: How was the fluorescence sensor on the CTD calibrated? Please provide full details in the methods section, including sample collection and treatment prior to extraction. If the sensor was calibrated then I am not sure I understand the need to re-calibrate the sensor output data with the Chl a concentrations determined here. The slope is less than 1 suggesting that pigments may have deteriorated using the approach outlined on P5, L 24-29 relative to the samples that were collected and extracted for calibration of the sensor. Providing further methodological details on the sample treatment and extraction process during Chl a extraction is needed, for example, were standards and internal reference materials treated in the same way as samples?

P8, L20: “showed” not “shows”

P8, L21: “concentration” should be plural

P8, L24: “were” not “are”

P8, L25: Suggest restructuring this sentence as follows: “POC and PN concentrations were highest near the coast on the inner shelf ($>90 \mu\text{g L}^{-1}$ and $>21 \mu\text{g L}^{-1}$, respectively), and decreased gradually with distance offshore (Fig. 4).”

P8, L27: Delete “nearby off” and insert “of” between “northeast” and “Taiwan”

P8, L28: Insert “by” between “varied” and “more”.

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

P8, L34: “Consistent with the POC distribution”

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

P9, L4: “was” not “is”

Discussion

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

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

P9, L22: Insert “at” between “except” and “these”

P9, L24: Suggest “...further delineated the area and water depths influenced by...”

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

P9, L30: Delete “to”

P10, Molar C/N Ratio: Do Wu et al (2003) and Liu et al (1998) see any cross shelf trends in C/N ratio? You quote considerably more variable C/N ratios from their studies than you have found in the DCM layers. Is this due differences in sampling depth, water mass, community composition, nutrient status, light status, age/residence time, degree of degradation, season, year etc.?

While I agree that there is likely a dominance of marine-POM in your DCM samples, I think a narrow range of C/N ratios alone does not provide enough evidence to confirm a lack of terrestrial signals transported mainly by the Yangtze River. Perhaps link these C/N ratios to salinity data presented in Fig. 2, which shows a greater influence of higher S waters and Fig. 7 which indicates a strong influence of KSSW across the shelf and minimal influence of the less saline CDW.

P11, POC/Chl a: I think it would perhaps be useful here to also note that both vertical gradients in community composition and photoacclimation can influence C/N and POC/Chl a within subsurface chlorophyll maxima.

Moore et al. (2006) Phytoplankton photoacclimation and photoadaptation in response to environmental gradients in a shelf sea, L&O DOI: 10.4319/lo.2006.51.2.0936

Latasa et al. (2017) Distribution of phytoplankton groups within the deep chlorophyll maximum. L&O, DOI: 10.1002/lno.10452.

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

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

P14, L30 – P15, L6: How would local regeneration of N and regenerated production influence these isotopic values, in addition to the $\delta^{15}\text{NO}_3$ of source waters?

P15, L17 – L33: I found this paragraph hard to follow and overly speculative.

“... may not be resulted from the high degree of nitrate utilisation, but the incorporation of inorganic nitrogen in the POM.” Nitrate is inorganic nitrogen, do you mean that there is a higher proportion of particulate inorganic nitrogen relative to particulate organic nitrogen and that this could be driving the isotopic signal here?

“The low Chl fluorescence might be limited by the low temperature in this high nutrient low chlorophyll region.” The depths where you sampled were quite warm and of similar temperature to one another, I think a temperature effect on Chl here is unlikely. These appear to be the deepest samples at ~100m. Is it possible that this signature of low Chl and low POM concentration is because you were sampling below the euphotic layer? This could possibly contribute to the isotopic signal at this station, as below the euphotic layer remineralisation and degradation of POM would exceed production. The $\delta^{13}\text{C}_{\text{DIC}}$ at depth may have been quite different to that in the euphotic layer. If dissolved inorganic nutrient data and PAR data are available and support your argument that it is a temperature effect then this should be included.

No NH_4^+ data are available, and no $\delta^{15}\text{N}_{\text{NH}_4^+}$ values have been previously published for this region, therefore I find the discussion linking the high $\delta^{15}\text{N}_{\text{PN}}$ to ammonium assimilation quite speculative.

P16, L5: insert “respectively” after “Wu et al”

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

P16, L10: “has been reduced”

P16, L12: “...reported that the particulate load discharged by...”

P16, L22: “Accompanying the decreasing...”

P17, L7: “...despite the study are being the best...”

P17, L9: As I understand it you did not measure primary production rates, you are inferring primary productivity from POC concentration. Therefore it may be more accurate here to say biomass rather than primary productivity. Again, to my knowledge you did not collect data on community

composition and therefore I am concerned that this statement is overly speculative for your concluding remarks.

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

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