‘Excess Nitrate’ in the East China Sea

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‘Excess nitrate’ was found in waters with salinities below 30.5 in the top 10–15 m of waters covering one-third to one-half of the East China Sea in the summer of 1992. In these waters, significant concentrations of [nitrate+nitrite], up to about 6 μM , could be found while the corresponding concentrations of phosphate remained at or below 0.07 μM . Thus, the ratio of [nitrate+nitrite] to phosphate in these waters was far in excess of that at which these two essential nutrients are utilized by marine phytoplankton. (‘Excess nitrate’ is the concentration of [nitrate+nitrite] in excess of that which may be utilized by marine phytoplankton at the observed concentration of phosphate.) As a result, in contrast to the open marine systems in general, primary production in a significant portion of the East China Sea may be phosphate-limiting rather than nitrogen-limiting. The area covered by this layer of combined nitrogen-rich water could be temporally variable. While no ‘excess nitrate’ was detected in the south-eastern East China Sea along the Chinese coasts in the summer of 1992, significant quantities of ‘excess nitrate’, up to 4 μM , were found at the same location in the early spring of 1993 in waters with salinities as high as 34.5.

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Introduction

The East China Sea is one of the larger and more productive marginal seas of the world. It extends from the Cheju Island, at about 33°20'N, in the north to the northern coast of the island of Taiwan, at about 25°N, in the south (Figure 1). It is bounded to the east by the Kuroshio and there is extensive exchanges between the East China Sea and the Kuroshio through frontal processes as well as the upwelling of the cold nutrient-rich deep Kuroshio water onto the shelf (Chern *et al.*, 1990; Su *et al.*, 1990; Wong *et al.*, 1991; Hsueh *et al.*, 1992; Liu *et al.*, 1992a,b; Chen *et al.*, 1995). As such, the East China Sea should be considered an open marine system. However, this sea is also bounded to the west by continental China from which it receives the outflow from Changjiang, one of the larger rivers of the world, at a rate of 979 km³ year⁻¹ (Milliman & Jin, 1985). This discharge of the Changjiang to the East China Sea is not evenly distributed throughout the year. It is at a minimum in the winter and it reaches a maximum in the summer (Beardsley *et al.*, 1985). The combination of the low discharge and the prevailing north-easterly wind in the winter confines the influence of the Changjiang to a narrow band along the coast. In the summer, under the combined effect of the high runoff and the

prevailing southerly winds, the Changjiang plume may propagate across the shelf and cover most of the northern part of the East China Sea (Beardsley *et al.*, 1985; Su & Weng, 1994). Hydrographic sections along the plume across the East China Sea frequently depict a tongue of fresher water extending from the mouth of the Changjiang seaward and it is underlain by a wedge of more saline water (Beardsley *et al.*, 1985). Thus, the East China Sea may behave like an estuarine system rather than a truly marine system and this may be especially true in the summer.

The Changjiang is an unusually combined nitrogen-rich river where nitrate concentrations as high as 70 μM (Edmond *et al.*, 1985) and N:P ratios exceeding 100 have been reported (Tang *et al.*, 1990). The annual net export of nitrate and phosphate to the East China Sea through the Changjiang have been estimated to be 60 $\times 10^9$ and 1.3 $\times 10^9$ mole year⁻¹, respectively (Edmond *et al.*, 1985). The vigorous turbulence and the associated high concentrations of suspended particulate material in the main channel of the inner Changjiang estuary limits photosynthetic activities in the mixing zone within the river so that biological removal of the nutrients from the Changjiang water occurs primarily in the open East China Sea rather than in its mixing zone (Edmond *et al.*, 1985). As a result, this nitrogen-rich water from

FIGURE 1. (a) The cruise tracks, station locations, bathymetry and the distribution of (b) salinity and (c) nitrate in the surface waters of the East China Sea in July, 1992.

the Changjiang may have been poured into the East China Sea with only minor modifications.

It is widely believed that primary production is phosphate- or P-limiting in the freshwater environment and nitrogen- or N-limiting in the marine and estuarine environment (Raymont, 1980) and that the relative availability of these two essential micro-nutrients may strongly affect the species composition of the phytoplankton in a body of water (Kilham & Hecky, 1988; Hecky & Kilham, 1988). However, although the East China Sea is an open marine

system, our results indicate that, unlike the open oceans, the surface waters covering a significant portion of this sea can be anomalously combined nitrogen-rich so that they may be P-limiting rather than N-limiting.

Sampling and analytical methods

Surveys

Stations were occupied in five transects, transects A-A' through E-E', in the East China Sea between 18

and 27 July 1992 aboard the R/V *Akademik Aleksandr Vinogradov* during Cruise V23/KM92 of the joint Kuroshio Edge Exchange Processes MArginal Seas Study (KEEP-MASS) of Russia and the Republic of China, and in one transect, transect T-T', between 10 and 17 July 1992 and again between 21 and 29 April 1993 aboard the R/V *Ocean Researcher I* during Cruise 323B and Cruise 352 B of the Kuroshio Edge Exchange Processes (KEEPKEY) Study. The cruise tracks and the station locations are shown in Figure 1.

Methods

At each station, the distribution of temperature and salinity were recorded with a SeaBird model SBE9/11 conductivity-temperature-depth (CTD) recorder. Discrete water samples were collected with GO-FLO bottles mounted onto a Rosette sampling assembly (General Oceanic). Sub-samples were returned to the shore-based laboratory for the determination of salinity, nitrate, nitrite and phosphate.

Salinity was measured with an Autosol salinometer. Sub-samples for the determination of the nutrients were quick-frozen with liquid nitrogen on board ship and stored frozen until they were analyzed in the laboratory. Nitrate and nitrite were determined by the standard pink azo dye method which has been adapted for use with a flow injection analyser (Morris & Riley, 1963; Strickland & Parsons, 1972; Gardner *et al.*, 1976; Pai *et al.*, 1990a; Liu *et al.*, 1992a,b). Phosphate was determined manually with the standard molybdenum blue method (Murphy & Riley, 1962; Strickland & Parsons, 1972; Pai *et al.*, 1990b). The precision for the determination of salinity, nitrate, nitrite and phosphate were ± 0.003 , ± 0.3 , ± 0.03 and $\pm 0.01 \mu\text{M}$, respectively.

Results and discussion

The bathymetry and the surface distribution of salinity and nitrate in the study area are shown in Figure 1. Saline surface waters are fed into the East China Sea primarily from the Kuroshio that forms the eastern boundary of the sea and secondarily through the Taiwan Strait as the Taiwan Strait Warm Current from the south (Chen *et al.*, 1995; Gong *et al.*, 1996). The salinities of these waters do not drop below 33.7 (Su & Weng, 1994; Chen *et al.*, 1995). Kuroshio water enters the East China Sea by frontal mixing as the warm Kuroshio Surface Water and by upwelling as the saline Kuroshio Tropical Water and the nutrient-rich Kuroshio Intermediate Water (Chen *et al.*, 1995). Thus, waters in the East China Sea with

salinities below about 34 would be seawater that has been modified by mixing with the incoming river water from the west. The isohaline at a salinity of 33 ran approximately diagonally from the southwest to the north-east of the study area across the shelf reaching beyond the 200-m isobath at the shelf break in the northernmost transect, transect E-E', and, dividing the East China Sea into approximately a fresher north-western two-thirds and a more saline south-eastern one-third. This indicates the extensive influence of the outflow from the Changjiang in the northern East China Sea in the summer.

The $1 \mu\text{M}$ -isopleth of nitrate coincided approximately with the isohaline of 30.5. This patch of fresher water with salinities below 30.5 and concentrations of nitrate as high as $6 \mu\text{M}$ was found in the north-western portion of the study area in transects D and E. Although some parts of the sea, notably the coastal waters off China, were not surveyed in this study, previous studies (Gu, 1990; Tang *et al.*, 1990) indicate that the concentrations of nitrate increases while salinity decreases shoreward towards the north and north-west of the sea and especially towards the mouth of the Changjiang. In this study, the isohaline of 30.5 extended from about 31°N and 127°E to about 26.5°N and 120.5°E . Areas of the sea, north and west of this isohaline would have been covered with waters with salinities below 30.5, and thus with concentration of nitrate above $1 \mu\text{M}$. If this extrapolation were true, waters with elevated concentrations of nitrate might have covered a third to a half of the sea. The corresponding concentrations of phosphate in these waters never exceeded $0.07 \mu\text{M}$. Thus, the ratio of nitrate to phosphate in these waters was considerably higher than the Redfield ratio (Redfield *et al.*, 1963) of 16 that is commonly found in marine waters and in marine phytoplankton (Redfield *et al.*, 1963; Hecky & Kilham, 1988). The presence of a residual amount of phosphate together with undetectable concentrations of nitrate in the surface waters within the euphotic zone has traditionally been used as a major line of evidence for N-limitation in marine waters (Raymont, 1980). In the freshwater environment, a N:P ratio substantially greater than 10 is considered to be indicative of phosphate-deficiency (Healey & Hendzel, 1980; Hecky & Kilham, 1988) that may limit primary production. If these criteria were applicable here, then, this patch of nitrate-rich surface water in the East China Sea was not N-limiting and might well have been P-limiting instead.

Since the surface concentrations of phosphate were frequently below the reliably measurable concentration of about $0.1 \mu\text{M}$, it is not meaningful to calculate the concentration ratio of nitrate to

phosphate in these waters. The enrichment in combined nitrogen in the study area is expressed, instead, as 'excess nitrate', or, $[\text{NO}_3]_{\text{ex}}$, such that

$$[\text{NO}_3]_{\text{ex}} = ([\text{NO}_3^-] + [\text{NO}_2^-]) - R[\text{P}]$$

where $[\text{NO}_3^-]$ and $[\text{NO}_2^-]$ are the concentrations of nitrate and nitrite, and $[\text{P}]$ is the concentration of inorganic phosphate. R is the ratio of the biological utilization of [nitrate+nitrite] relative to phosphate by marine phytoplankton. Thus, in N-limiting waters where a residual amount of phosphate is present while nitrate is undetectable, there will be a negative 'excess nitrate'. On the other hand, in P-limiting waters, there will be a positive 'excess nitrate' which represents the amount of [nitrate+nitrite] that will remain if phosphate is being depleted by biological utilization. [In the study area, when the concentration of nitrate exceeded $1 \mu\text{M}$, the concentration of nitrite was invariably much lower than that of nitrate. Thus, [nitrate+nitrite] was almost identical to nitrate. Although ammonia and organic nitrogen were not measured in this study, their concentrations in this general area have been shown to be much lower than that of nitrate in previous reports (Edmond *et al.*, 1985; Sagi, 1990)]. A value of 14.5 was used for R in this calculation since the nitrate to phosphate ratio in the Kuroshio water that may find its way onto the shelf is at approximately this value (Wong *et al.*, 1989; Liu *et al.*, 1989; Gong, 1992). The estimated concentrations of 'excess nitrate' in the surface waters did not change appreciably for values of R within the probable range of 14–16 since the concentrations of phosphate in these waters were rather low. In the deeper waters, the concentration of phosphate was high enough so that the estimated concentration of 'excess nitrate' could be quite sensitive to small changes in R . Thus, the distribution of 'excess nitrate' in the deeper waters should be interpreted with caution.

The distribution of salinity, $[\text{NO}_3]_{\text{ex}}$, and nitrite in the surface waters in the northernmost transect, E-E', across the shelf and in an along shelf transect M-M', which linked the stations with the lowest surface salinity in each cross-shelf transect, are shown in Figure 2. 'Excess nitrate' as high as about $6 \mu\text{M}$ was found in the waters with salinities below 30.5. High concentrations of nitrite were found concomitantly with the high concentrations of 'excess nitrate' in both transects. Nitrite may be formed as an intermediate during nitrification or nitrate reduction (Spencer, 1975). It is unclear, from this data set alone, what the source of this nitrite may be and whether it may have any biogeochemical significance in relation to the

dynamics of the utilization of the nutrients in this unique type of water.

Aside from the outflow from Changjiang, the other possible major sources of water to the East China Sea are of marine origin and they are the Kuroshio Surface Water, the Kuroshio Tropical Water, the Kuroshio Intermediate Water and the Taiwan Strait Warm Current. Since, in the computation of 'excess nitrate', nitrate of marine origin was corrected for, all waters with a marine origin would have zero 'excess nitrate'. Furthermore, with the exception of the upwelling waters, the correction was small since the concentrations of the nutrients in these surface seawaters were low. Thus, the relationship between 'excess nitrate' and salinity may be treated as a two end-member mixing diagram between river water and seawater. The relationship between 'excess nitrate' and salinity between salinity 25 and 33 is shown in Figure 3. Although our data did not cover salinities below 25, it was obvious that the data points fell below the conservative mixing line (between $S=35$, $[\text{NO}_3]_{\text{ex}}=0 \mu\text{M}$ and $S=0$, $[\text{NO}_3]_{\text{ex}} > 6 \mu\text{M}$). Since 'excess nitrate' reached zero at a salinity of about 31, it was completely utilized in waters with salinities below this value. The salinity at which 'excess nitrate' may become undetectable will depend on the amount of 'excess nitrate' that is added to the Sea, the intensity of biological activity which controls the rate of utilization of 'excess nitrate' in the fresher waters in the sea, and the mixing intensity between the incoming river water and seawater. These factors may of course vary with time and space.

As a first approximation, between salinities 25 and 31, the relationship may be represented by the following linear equation:

$$[\text{NO}_3]_{\text{ex}} = -1.04(\pm 0.14)S + 31.7(\pm 1.1) \quad r=0.82$$

Thus, 'excess nitrate' decreased with increasing salinity and it became undetectable at a salinity of 30.5 so that this isohaline may be used to mark the area covered by this combined nitrogen-rich water. The sections of salinity and 'excess nitrate' along transect E-E' are shown in Figure 4. The waters with salinities below 30.5 and a detectable amount of 'excess nitrate' formed a tongue across most of the transect all the way to the shelf edge and down to about 15 m. Thus, if indeed the presence of 'excess nitrate' signifies P-limitation, since waters with salinities below 30.5 not only covered about a third to a half (Figure 1) of the East China Sea as discussed previously but also penetrated to a depth of 10–15 m (Figure 4) in the summer, P-limitation might have been a rather

FIGURE 2. The distribution of salinity (open circle), 'excess nitrate' (filled circle) and nitrite (open triangle) in the surface waters along (a) E-E' in July, 1992; (b) M-M' in July, 1992; and (c) T-T' in April, 1993.

prevalent condition in this sea. The intercept of the linear line relating 'excess nitrate' to salinity (Figure 3) suggests that 'excess nitrate' at zero salinity was only 32 μM , a concentration that is considerably lower than that of nitrate found in the Changjiang. If little [nitrate+nitrite] is removed within the Changjiang estuary and the concentration of nitrate in Changjiang water is on the order of about 70 μM (Edmond *et al.*, 1985), this would imply that about half of these nitrogen species has already been removed as the Changjiang plume travels from the river mouth to the study area across the inner shelf of the East China Sea. Although the relationship between 'excess nitrate' and salinity between salinity 25 and 31 was approximately linear, there was considerable scatter. In fact, the data points from sections D-D' and E-E' may have fallen on somewhat different lines (Figure 3). These variabilities may be explained by temporal variations in the composition of Changjiang water and/or the

removal and/or regeneration of the nitrate and nitrite within this salinity range. At the most landward stations, there was a pool of bottom water with slightly negative 'excess nitrate' in section E-E' (Figure 4). This pool of water has been identified as modified Yellow Sea Cold Water in a more detailed analysis of the hydrography observed during this cruise (Gong *et al.*, 1996). This would suggest that Yellow Sea Cold Water was enriched in nitrate and nitrite relative to phosphate. Nonetheless, the concentration of negative 'excess nitrate' found was quite small and may not be reliable enough for defining another water mass.

Although there was no detectable amount of 'excess nitrate' in the southern-most transect, T-T', during July 1992, when that transect was re-sampled in the early Spring, in April 1993, 'excess nitrate', as high as 4 μM , was found at the two landward-most stations and the high concentrations of 'excess

FIGURE 3. The relationship between 'excess nitrate' and salinity in transects E-E' (solid circle), D-D' (open circle), B-B' (solid triangle), C-C' (open triangle), A-A' (solid square) and T-T' (open square) at salinities below 33. Solid line denotes the linear least square fit of all the data points with salinities below 31.

nitrate' were again accompanied by high concentrations of nitrite and lower salinities [Figure 2(c)]. This distribution of 'excess nitrate' is consistent with the concept of an intensified southerly flow of the water from the Changjiang along the coast in the winter and suggests a temporal variation of the area of the East China Sea that is covered by the nitrogen-rich and possibly P-limiting water. The surface salinities at the two stations where 'excess nitrate' was found

were between 32.5 and 34.5. These salinities were significantly higher than those in which 'excess nitrate' was found in summer, 1992. Thus, 'excess nitrate' could be found in more saline water, at least up to a salinity of 34.5, in the winter and early spring. If this relationship between 'excess nitrate' and salinity is indicative of what happens in the entire winter, it would mean that even under low flow, waters with 'excess nitrate', that is, waters potentially under P-limitation, may still cover a significant portion of the East China Sea since waters with salinities below 34.5 is quite commonly found in this sea in the winter (Gu, 1990). As a result, the presence of waters with 'excess nitrate' in a significant portion of the East China Sea may be a year round phenomenon although it may be most prevalent in the summer.

The presence of waters with anomalously high ratios of nitrate to phosphate in the East China Sea has been observed in previous studies (Sagi, 1990; Tang, 1990). However, it was noted only in passing and its significance has not been considered. Given the recognized effect of the relative availability of phosphate and nitrate on the biological makeup of a body of water (Hecky & Kilham, 1988; Kilham & Hecky, 1988) and the limitation of phytoplankton growth in marine systems by the availability of nitrogen (Raymont, 1980), the East China Sea may not behave like a typical marine system. The ramifications of this peculiar characteristic of the East China Sea on its biogeochemistry should be and have yet to be further explored. The Chinese government is in the process of building a series of dams across the Changjiang. Once the dams are completed, the outflow to the East China Sea will be greatly reduced and the phenomenon reported

FIGURE 4. The distribution of (a) salinity and (b) 'excess nitrate' in transect E-E'.

here may become much less extensive or even absent. The resulting impact on the biogeochemical characteristics of the East China Sea is unknown but is likely to be significant.

Conclusions

The East China Sea is enriched in combined nitrogen. Relative to the concentration of phosphate, [nitrate+nitrite] was present in excess of the amount needed to support phytoplankton growth in waters covering the north-western one-third to one-half of the sea, down to a depth of 10–15 m in the summer of 1992. There was also an indication that this phenomenon may be found during other seasons of the year in a significant portion of this sea. The presence of this 'excess nitrate' suggests that, unlike other marine systems, the East China Sea may be P-limiting rather than N-limiting.

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