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A Sediment Chronology of the Eutrophication of Chesapeake Bay

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ABSTRACT: Chesapeake Bay sediments were examined for biogeochemical evidence of eutrophication trends using two mesohaline sediment cores. Measurements of ²¹⁰Pb geochronology and sediment profiles of organic carbon, nitrogen, organic phosphorus, inorganic phosphorus, and biogenic silica (BSi) were used to develop temporal concentration trends. Recent sediments have 2–3 times as much organic carbon and nitrogen as sediments from 80 to 100 yr ago, but the increases result from both changes in organic matter deposition and time-dependent changes in organic matter decomposition rates. Despite increases in phosphorus loading, no major changes in phosphorus concentration were noted throughout most of the century; anthropogenic phosphorus deposition, though not evident in sulfidic mid-bay sediments, must occur in more oxidizing sediment environments in both the northern and southern bays. Temporal trends in BSi concentrations are much less evident and the lack of substantial increases in this century suggest that BSi inputs may be capped by late spring-summer Si limitation.

Introduction

Temporally and spatially variable inputs of pollutants into aquatic ecosystems are best assessed by careful measurement of individual point source and nonpoint source inputs. Such detailed studies provide critical information needed to choose effective management strategies for minimizing coastal pollution. For most aquatic systems, such detailed information is lacking for both logistical and monetary reasons. Although individual pollutant sources can be hard to identify, sediment geochronological studies have provided temporal trend information on contaminant inputs to aquatic systems (Livett 1988; Valette-Silver 1993). Heavy metals and organic contaminants often have a strong affinity for particle surfaces, and in many cases, dated sediment profiles can provide an accurate chronology of contaminant inputs. A number of basic assumptions go into both the core dating process and the consequent estimation of contaminant inputs; in many cases, such geochronological reconstruction is not possible.

Determining the time course of nutrient input and cycling within aquatic systems using dated cores is more difficult than determining the inputs of heavy metals because a large proportion of N, P, and C can be recycled to the water column rather than preserved in the sediment (Nixon 1981; Boynton et al. 1994), and the overall rates and pathways of the recycling of nutrient elements may change with eutrophication (e.g., Kemp et al. 1990). Sediment chronologies of nutrient inputs to lacustrine ecosystems have proven valuable to understanding the time scales of eutrophication in the Laurentian Great Lakes (Kemp et al. 1976; Conley et al. 1993) and other lakes (Edmondson 1974). Estuarine systems are fundamentally more difficult systems for such studies because of stronger physical forcing, increased spatial variabilities, and both enhanced rates and increased depths of sediment bioturbation (Nixon 1988). In this paper, we examine the utility of using a sediment nutrient chronology to provide insight into eutrophication of the Chesapeake Bay over the last 100 yr.

The Chesapeake Bay has experienced serious environmental degradation in the last half of the 20th Century. Inputs of nitrogen and phosphorus from the watershed have increased significantly, resulting in higher dissolved nutrient concentrations and phytoplankton biomass (Harding 1994). Responses to high nutrient inputs and increased planktonic primary production include a loss of most submerged vascular plants (Kemp et al. 1983; Orth and Moore 1984), a decrease in the impor-

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Fig. 1. Map of Chesapeake Bay station locations. Station M3 is located on the western flank of the deep channel in 15 m of water. Station HI is located at the confluence of the Patuxent River and the main stem of the Chesapeake Bay in 17.5 m of water.

tance of benthic algal production (Brush and Davis 1984; Cooper and Brush 1993), and an increased extent of anoxia in mesohaline bottom waters (Smith et al. 1992). Cooper and Brush (1991) have documented the long-term (2,500 yr) changes in organic C and organic N concentrations in Chesapeake Bay mesohaline sediments, but temporal resolution at the time of accelerated eutrophication in the 1900–1980 period was limited.

In this paper we utilize chronologies of nutrient element (C, N, P, Si) burial at two Chesapeake Bay mesohaline sites in order to address two questions: Are temporal changes in nutrient inputs reflected in the nutrient element sediment chronology? and What was the timing of the eutrophication of the bay? This study is the first sediment nutrient study in the Chesapeake Bay with an emphasis on recent (<50 yr) eutrophication trends within the estuary.

Methods

The mesohaline Chesapeake Bay was chosen for core collection because the region below the turbidity maximum zone is characterized by extremely high primary production, because water column and sediment nutrient studies in the past decade have provided a first-order understanding of biogeochemical processes in this region (i.e., Smith et al. 1992), because bioturbation is minimized by

seasonal anoxia (Reinharz et al. 1982), and because this region has been the site of other successful sediment dating efforts (Officer et al. 1984; Dibb and Rice 1989). Of 10 cores collected for ²¹⁰Pb dating in the mesohaline Chesapeake Bay, two were selected for a detailed chronology of nutrient burial. These two cores were chosen because 1) seasonal anoxia limits the potential for bioturbation, and 2) the measured sedimentation rates of ~ 0.5 cm yr⁻¹ provide excellent temporal resolution for the past ~ 100 yr in the 50–70 cm cores that are readily obtained using conventional box corers. Site M3 (38°34.05'N; 76°26.76'W) is located just south of the Choptank River in 15 m of water on the western flank of the main channel (Fig. 1). This site has been intensively studied for temporal changes in porewater and solid-phase chemistry, sediment metabolism, nitrogen cycling, and sediment-water exchange as part of the Land Margin Ecosystem Research Program (LMER Coordinating Committee 1992). Site HI (38°18.47'N; 76°18.69'W) is located immediately off the Patuxent River about 20 km south of site M3 in 17.5 m of water.

A Soutar box corer was used to collect a 51 cm core at site HI and a University of Delaware box corer was used to collect a 70 cm core from site M3. Both cores appeared undisturbed, with an in-

tact flocculent surface layer. The box cores were immediately subcored using a 15-cm diameter acrylic tube and sediment samples were extruded within 2 h of collection. The sediment was sectioned into intervals of 0.5 cm, 1.0 cm, or 2.0 cm. Sediments were homogenized, split into samples for nutrient, sulfur, and contaminant analysis, and frozen. Samples for nutrient and ²¹⁰Pb analysis were dried at 65°C and ground with a mortar and pestle.

The measurement of ²¹⁰Pb ($T_{\frac{1}{2}} = 22.3$ yr) was carried out by the analysis of its daughter radionuclide, ²¹⁰Po ($T_{\frac{1}{2}} = 138$ d). Samples were stored for several ²¹⁰Po half-lives to ensure secular equilibrium between the two nuclides. The extraction procedure for ²¹⁰Po generally followed that of Flynn (1974), employing some of the modifications by Benoit and Hemond (1988). The unsupported ²¹⁰Pb activity was determined from the asymptote of the ²¹⁰Po profiles. Cumulative mass (g cm⁻²) was converted from percent water data using a sediment density of 2.5 g cm⁻³. The constant initial concentration sedimentation model was used (Robbins 1978) and a linear regression of unsupported ²¹⁰Pb versus cumulative mass was used to provide a sedimentation rate in $g \text{ cm}^{-2} \text{ yr}^{-1}$. The age at a given sediment depth was estimated by dividing the cumulative mass by the sedimentation rate.

The analysis of total carbon and nitrogen utilized a Control Equipment CHN analyzer. Total and inorganic P was extracted from ashed and unashed sediments using 1 N HCl, with subsequent colorimetric analysis (Aspila et al. 1976). Inorganic carbon was determined by acidification and analysis of CO_2 by gas chromatography (Stainton 1973). Organic carbon was calculated as the difference between total and inorganic carbon. Biogenic silica (BSi) was analyzed via a time series digestion using Na₂CO₃ (DeMaster 1981). Aluminum was analyzed by a fusion technique followed by atomic absorption spectrophotometry (Medlin et al. 1969).

Results

The ²¹⁰Pb profiles in sediment cores M3B and HI generally follow an exponential decrease, with an apparent mixed layer of several cm (Fig. 2). The regression model gives an excellent fit for core M3B and a somewhat poorer fit for core HI (Fig. 2). Despite differences in the shape of the ²¹⁰Pb profiles, the age at a given core depth is virtually identical for these cores. The predicted sedimentation rates were 1,800 g m⁻² yr⁻¹ and 2,400 g m⁻² yr⁻¹ for cores M3B and HI respectively, rates similar to those obtained for other studies in the Chesapeake Bay mesohaline region $(1,000-3,000 \text{ g m}^{-2} \text{ yr}^{-1}; \text{ Officer et al. 1984; Cooper and Brush 1993}).$

Vertical profiles of organic C, total N, and total P for both cores M3B and HI followed similar trends, generally showing enrichment in more recent sediments (Fig. 3). Organic carbon and nitrogen concentrations increased smoothly by a factor of 2 or more from deep to surface sediments. For both organic C and N, there are consistent subsurface peaks, with slightly lower concentrations in the top 1–2 cm. Profiles of BSi concentration were more varied, with M3B showing a recent increase from ~40 mg g⁻¹ below 10 cm to almost 60 mg g⁻¹ in near-surface sediments. BSi concentrations in core HI showed higher variability, with sediment concentrations at 30 cm generally lower than those found near the surface and at depth.

While the increases in P concentrations have similarities to those of C and N, below 20 cm the P concentrations are constant, increasing abruptly in near-surface horizons (Fig. 3). Core HI has the sharpest increase, the concentration almost doubling in the top 5 cm. Analysis of organic versus inorganic components of sediment P provides more insight into the cause of the P enrichment (Fig. 4). Near-surface enrichment of P in core HI results primarily from inorganic P enrichments while those in core M3B arise from organic P increases. Such inorganic enrichments have been observed at M3B during fall, winter, and spring, but they are remobilized from sediment during summer anoxia, such as that occurring at the time of collection of core M3B (Cornwell and Owens unpublished data); bottom waters were not anoxic when core HI was collected.

The sharpest increases in nutrient concentrations occurred in sediment horizons corresponding to-the 1980s (Fig. 5). The profiles of total N at M3B and HI are similar. The profiles of organic carbon are not as smooth as those for total N, particularly in the 1950–1970 horizons. In contrast to the C and N profiles, increases in total P concentrations occur only in recent years.

Discussion

ORGANIC CARBON AND NITROGEN

Although it is tempting to associate temporal increases in sediment organic carbon and nitrogen concentrations to eutrophication, they may actually result from a combination of depositional and postdepositional processes. The observed increases in organic matter concentrations in more recent sediments can result from several factors including a steady-state input-decomposition balance, with surficial sediments having higher organic carbon because less of the labile portion has decomposed;



Fig. 2. Sediment ²¹⁰Pb data. Total ²¹⁰Pb activity for M3B and HI are shown in the upper panels; plots of the natural log of excess ²¹⁰Pb versus cumulative mass are shown in the lower panels. Linear regression was used to estimate the slope and allow calculation of the sedimentation rate.

increased inputs of organic matter to the sedimentwater interface; and altered decomposition pathways and sediment organic matter preservation associated with changing redox conditions. Each of these potential causes must be evaluated in order

to interpret the observed organic carbon profiles in terms of implications for the history of eutrophication in Chesapeake Bay.

To evaluate the question of whether depositional or diagenetic processes shape the organic carbon



Fig. 3. Sediment nutrient profiles for organic carbon, total nitrogen, total phosphorus, and biogenic silica in cores M3B and HI.

profiles, it is useful to examine the profiles predicted for a system with constant organic matter inputs. Organic matter decomposition may be modelled using a variety of models, which generally utilize first-order kinetics (Berner 1964): where G is the labile organic matter pool, t is time, and k is a first-order rate constant. Simple firstorder decomposition models are valid over short time periods; for longer time periods, several different pools of labile organic matter are assumed, all with different rate constants of decomposition

$$dG/dt = -kG \tag{1}$$

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Fig. 4. Sediment phosphorus profiles. Total and organic phosphorus concentrations are presented; the difference between the organic and total phosphorus concentrations in each sample is inorganic phosphorus.

(Berner 1980; Westrich and Berner 1984). Middelburg (1989) evaluated the use of a simple model with a time-dependent decomposition rate parameter:

$$k = 0.16t^{-0.95}$$
(2)

Solving Eq. 1 with the time-dependent k from Eq. 2, the concentration of total organic carbon can be estimated as a function of time (Middelburg 1989):

$$G_{t} = G_{0} \exp(-3.2t^{0.05}) \tag{3}$$

Burdige (1991) has noted that decomposition constants decreased with depth in southern Chesapeake Bay sediments. The model output (Fig. 6) shows a rapid decrease in organic matter concentrations in the first few years of decomposition and relatively slow rates of decomposition after several decades. The value of G_0 was chosen to provide general correspondence between the model and near-surface organic carbon concentrations. The purpose of showing the model output in Fig. 6 is not to calibrate the Middelburg model, but rather to show that much of the observed change in organic matter concentrations can be accounted for by steady-state diagenesis. The overall correspondence of the organic carbon model prediction with that of the data is relatively good except in the middle part of the century where the model underestimates the organic carbon concentrations. Clearly, the organic matter profiles from the 1970s to the time of core collection are shaped strongly by decomposition, while the profile prior to the 1970s partially reflects an increase in deposition.

The textural characteristics of sediment can also have an influence on the concentration of organic matter. Mayer (1994) has shown that the surface area of particulates is a prime determinant of the organic matter that is preserved in sediments. Normalization of trace metal and nutrient concentrations to Al has been used extensively for correcting for changes in grain size (Finney and Huh 1989).



Fig. 5. Time courses of nutrient concentrations in cores M3B and HI.



Fig. 6. Organic concentration versus time for cores M3B and HI. The bottom panel shows the same organic carbon data normalized by dividing it by the concentration (mg g^{-1}) of total aluminum. The solid line is the output of the Middelburg (1989) diagenetic model of organic matter decomposition; initial values of organic carbon were selected to match the data at the surface and at depth.

In Fig. 6, the organic carbon concentrations are expressed as a C:Al ratio. The overall pattern of normalized concentration is similar to that found for organic carbon concentrations, though the proportional range increases in the normalized case. Lower Al concentrations in the top few core sections result in high C:Al ratios. Grain size does not appear to be a major cause of changes in the organic carbon profile.

An alternative explanation for increased organic matter concentrations in near-surface sediments could be changes in carbon preservation rates associated with changes in the duration and degree of oxygen depletion. A number of studies have suggested that the extent of anoxia has increased during this century (Smith et al. 1992); the modern extent of anoxia appears to be controlled by both physical processes (such as spring freshwater inflow) as well as increased nutrient inputs in high flow years (i.e., Boicourt 1992). Recent evidence suggests anoxia may not affect long-term preservation rates of organic C (Henrichs and Reeburgh 1987; Lee 1992).

Evidence for increased organic C inputs has been found in relatively few estuarine and coastal systems. Increases in organic matter deposition have been associated with sewage inputs of particulate N (Stull et al. 1986) and organic C (Finney and Huh 1989) to coastal waters near Los Angeles, California. Nixon (1988) has noted that, relative to lacustrine sediments, there is a general absence of sedimentary evidence for long-term increased inputs of organic C, N, and P in estuarine sediments. He attributes such lack of evidence at least partly to increased physical mixing rates.

BIOGENIC SILICA

Variation in BSi concentration in sediment profiles is often used to infer productivity of overlying waters. For example, in the North American Great Lakes, increases in BSi accumulation have been related to eutrophication that results from increases in P loading rates into strongly P-limited systems (Conley et al. 1993). The lack of substantial increases in BSi concentration in Chesapeake Bay sediments is somewhat puzzling given the environmental degradation in the last half of the 20th Century. However, over much of this century, dissolved silicate supplies during the spring have been completely utilized in the Chesapeake Bay during the winter-spring diatom bloom (Conley and Malone 1992). Further increases in BSi accumulation, therefore, may be limited because the system fully utilizes its dissolved silicate supplies in the estuary on an annual basis; further increases in BSi accumulation could not occur under this scenario.

PHOSPHORUS

Phosphorus profiles in aquatic sediments are shaped by depositional and diagenetic processes. Postdepositional mobility of inorganic P can result in the flux of P across the sediment-water interface (Nixon 1981; Caraco et al. 1989) or the redistribution of solid-phase inorganic P (Carignan and Flett 1981; Cornwell 1987). Of concern for an interpretation of past P deposition are 1) temporal changes in the efficiency of inorganic P burial and 2) formation of P-enriched sediment horizons under more oxidizing sediment conditions. The steady increases during the mid-century in organic C and N are not found for total P, inorganic P, or organic P (Fig. 5). The recent increases in P concentration in the sediment correspond to the time

of rapid increases in sediment organic C and N. The large increases in inorganic P at station HI represent a diagenetic enrichment or a temporary persistence of Fe-associated inorganic P in more oxidized surficial sediments (i.e., Krom and Berner 1981). The P data from these cores do not show any long-term temporal changes in P inputs, only recent enrichment that may decrease as a result of dissolution of Fe-P complexes or the decomposition of P in organic matter.

CHANGES IN SEDIMENT ORGANIC MATTER COMPOSITION

Elemental ratios of organic matter provide useful indices of sediment provenance (i.e., terrestrial versus estuarine) and differential rates of sediment decomposition for C, N, P, and BSi (Fig. 7). The profiles of C:N ratios varied greatly but were closest to Redfield ratios (~ 6.6) in near-surface sediments, where ratios were similar to those observed previously for both seston and surficial sediment (Boynton and Kemp 1985) in the mid-bay region. Peak C:N ratios were found mid-century in both cores, with the greatest amplitude found in core M3B. The source of N depletion (relative to organic carbon) is not readily apparent. Enhanced decomposition rates for N can account for the increase in C:N ratio as sediment ages; the subsequent decrease to lower values early in the century is not consistent with this explanation. It would not be expected that high C:N sewage-derived particulates would be important in this part of the Chesapeake Bay and increased fluvial inputs of high C: N terrestrial particulates are unlikely in this midbay region where sediment inputs are dominated by shoreline erosion (Hobbs et al. 1992). Anthropogenic inputs of N-depleted organic matter from coal, fossil fuel combustion by-products, or oil pollution have the potential to increase the C:N ratio, though no direct measurements are available at these sites. Detailed chemical analysis to determine the forms of organic C and N in the sediments is necessary to decipher these C:N ratio profiles.

Organic C:total P ratios generally range from ~ 100 to ~ 150 , with higher values in mid-century. Values most similar to Redfield ratios are found in very recent sediments and in very old sediments. In general, there has been a trend of increased organic C:total P ratios, with the exception of near-surface sediments. This increase in organic C:total P ratios occurs in sediment that was deposited when the anthropogenic inputs of P were increasing rapidly. Ingall and Van Cappellen (1990) have shown that for marine sediments, organic C:organic P ratios above 200 are typical for sediments with accretion rates between about 0.002 cm yr⁻¹ and 1 cm yr⁻¹, a range encompassing the 0.5 cm yr⁻¹

found at these two sites. Organic C:total P ratios are much higher than Redfield ratios, reflecting the more complete decomposition of organic P relative to C. Mid-century sediments from core M3B are enriched in organic C relative to organic P.

The N:total P ratio increases throughout the century, with ratios similar to the Redfield ratio near the sediment-water interface. This may be partly a diagenetic effect because some remineralized P may precipitate in oxidized sediments near the interface. This mechanism is not available for the retention of N, and remineralized N generally is transported across the sediment water-interface as a solute or gas (i.e., Kemp et al. 1990) rather than stored in an inorganic form.

NUTRIENT LOADING TRENDS IN CHESAPEAKE BAY

Nutrient inputs into the Chesapeake Bay have a variety of point and diffuse sources. Boynton et al.'s (1995) N and P budgets show that point sources compose roughly ¹/₃ of total inputs, with the rest coming in primarily through surface water and groundwater inputs. While it is difficult to quantify the changing inputs of nutrients into the Chesapeake Bay, increasing population and more intensive applications of inorganic fertilizer and manure greatly increased nutrient inputs into the Chesapeake Bay in this century. Such trends are illustrated in Fig. 8, which shows fertilizer use tripling in the state of Maryland in the time period 1940-1980. Because of the importance of nonpointsource nutrient inputs, even a doubling of such diffuse sources would have a major impact on the nutrient budget. In the Potomac subestuary, Champ et al. (1981) estimated that in the time period 1913–1970 when the population increased by a factor of 8, total N and total P inputs from wastewater in the greater Washington, D.C., area increased about 10 and 20 times, respectively. Furthermore, Baltimore did not have a sewage collection system until 1906, so point-source inputs there were not a factor at the turn of the century. It would be easy to hypothesize at least a tripling of nutrient inputs from 1900 to the 1970s.

EVIDENCE FOR EUTROPHICATION

This examination of the sediment biogeochemical evidence for changing nutrient inputs to Chesapeake Bay is confounded by postdepositional changes in nutrient element concentrations. Although the high concentrations of organic C and N in sediments <20 yr old are consistent with enrichment with more labile organic matter, increasing organic matter concentrations found in sediments during the first 60 yr of the 20th Century suggest there has been substantially increased or-



Fig. 7. Changing elemental ratios for nutrient elements. A large mid-century increase in the molar carbon: nitrogen ratios is evident in cores M3B and HI. Steady increases in the organic carbon to total phosphorus ratio and nitrogen: total phosphorus ratio in this century indicate lower preservation of phosphorus relative to nitrogen and carbon.



Fig. 8. Annual fertilizer use in the state of Maryland during the period 1935–1985. The first documented use of inorganic fertilizers in Maryland was in 1824.

ganic matter loading. This conclusion is similar to that of Cooper and Brush (1991).

The absence of increased P deposition when P loading increased throughout this century results from geochemical remobilization of P from sediments. Under anoxic conditions, P retention by deep-water mesohaline sediments is limited by the surface area of iron oxides, which adsorb inorganic P. Current budgets of P for the Chesapeake suggest that Chesapeake Bay imports P from the coastal ocean, rather than exporting it (Boynton et al. 1995). Consequently, increases in P loading must result in increased P deposition. In the Baltic Sea, Carman and Wulff (1989) have shown that, relative to anoxic deep-water sediments, there is increased sediment P adsorption capacity and deposition in shallow water, more oxidizing sediments. This must also be the case for the Chesapeake Bay, with increased P deposition in sedimentary regimes other than below the pycnocline in the mesohaline region. Obtaining sedimentary evidence for increased P deposition in such shallow water is confounded by the activity of bioturbating organisms.

Despite large increases in nutrient loading, there does not appear to have been commensurate increases in N and P deposition in these mesohaline sediments. While P may experience enhanced retention in sediments of high adsorption capacity, this does not explain why there has not been a stronger response in these sediments to increases in N loading. A nonlinear response of deposition to overall system loading requires other processes to explain the response. In the Chesapeake, other sinks for N and P might include benthic algae in shallow water, submerged aquatic vegetation, finfish, and shellfish. Major alterations in the distribution and biomass of living resources have occurred during this century in the Chesapeake Bay, and these changes may have not only been a response to nutrient loading, but also a factor altering the pathways of nutrient cycling within the system.

The sites selected for this study were chosen because it was thought that they would provide the best record of changing nutrient inputs and eutrophication in the Chesapeake. This study shows that even in estuarine systems uncomplicated by mixing processes, the simple interpretation of nutrient element profiles may provide an inaccurate picture of temporal changes in nutrient loading.

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