

Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: Evolving views over three decades

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Abstract

The first special volume of *Limnology and Oceanography*, published in 1972, focused on whether phosphorus (P) or carbon (C) is the major agent causing eutrophication in aquatic ecosystems. Only slight mention was made that estuaries may behave differently from lakes and that nitrogen (N) may cause eutrophication in estuaries. In the following decade, an understanding of eutrophication in estuaries proceeded in relative isolation from the community of scientists studying lakes. National water quality policy in the United States was directed almost solely toward P control for both lakes and estuaries, and similarly, European nations tended to focus on P control in lakes. Although bioassay data indicated N control of eutrophication in estuaries as early as the 1970s, this body of knowledge was treated with skepticism by many freshwater scientists and water-quality managers, because bioassay data in lakes often did not properly indicate the importance of P relative to C in those ecosystems. Hence, the bioassay data in estuaries had little influence on water-quality management. Over the past two decades, a strong consensus has evolved among the scientific community that N is the primary cause of eutrophication in many coastal ecosystems. The development of this consensus was based in part on data from whole-ecosystem studies and on a growing body of evidence that presented convincing mechanistic reasons why the controls of eutrophication in lakes and coastal marine ecosystems may differ. Even though N is probably the major cause of eutrophication in most coastal systems in the temperate zone, optimal management of coastal eutrophication suggests controlling both N and P, in part because P can limit primary production in some systems. In addition, excess P in estuaries can interact with the availability of N and silica (Si) to adversely affect ecological structure. Reduction of P to upstream freshwater ecosystems can also benefit coastal marine ecosystems through mechanisms such as increased Si fluxes.

Nitrogen (N) pollution has increased remarkably over the past several decades as a result of increased creation of reactive N for fertilizer use and, inadvertently, from combustion of fossil fuels (Galloway et al. 2004). Both a “white-paper panel” of the Ecological Society of America (Vitousek et al. 1997) and the Coastal Marine Team of the National Climate Change Assessment (Boesch et al. 2000; Scavia et al. 2002) concluded that N pollution is one of the greatest consequences of human-accelerated global change on the coastal oceans of the world. The global distribution of reactive N is far from uniform, and N pollution in coastal waters is greatest where agricultural activity and urbanization are greatest. In some regions, such as the North Sea and Yellow Sea, human activity probably has increased N fluxes to the coast by a 10- to 15-fold measure or greater, while in other areas, such as Hudson’s Bay and Labrador, human activity probably has had little effect on N fluxes (Howarth 2003). On average for the United States, human activity probably has increased N fluxes to the coast by an estimated sixfold measure (Howarth et al. 2002; Howarth 2003), and some two thirds of the nation’s coastal waters are moderately

or severely degraded from N pollution, which is causing extensive eutrophication (Bricker et al. 1999). Today there is a scientific consensus, which has emerged from research at several spatial and temporal scales, that N represents the largest pollution problem in the nation’s coastal waters and one of the greatest threats to the ecological functioning of these ecosystems (Nixon 1995; Howarth et al. 2000b; NRC 2000). The societal implications are immense, as estuaries are among the most valuable of all ecosystems with regard to the services they provide, particularly for fish and shellfish production, recreation, and waste assimilation (Costanza et al. 1997).

Even in the early part of the 20th century, some scientists were suggesting that primary productivity in marine ecosystems was N limited (Johnston 1908), and by mid-century, Ryther (1954) provided some evidence for N rather than phosphorus (P) control of eutrophication in Moriches Bay on Long Island; see Nixon et al. (1986) for a review of this and other historical work on N in estuaries and coastal marine ecosystems. By the early 1970s, further evidence had accumulated that coastal eutrophication was caused primarily by N (Ryther and Dunstan 1971; Vince and Valiela 1973), and this viewpoint has been widely shared by many coastal marine scientists since the 1980s (Boynton et al. 1982; Granéli et al. 1990; Nixon 1995). However, N enrichment as the primary cause of eutrophication in estuaries and coastal seas was far from universally accepted over much of this time. Some limnologists argued that P was more likely to regulate primary production and eutrophication in estuaries, as in lakes (Hecky and Kilham 1988; Hecky 1998; Hellstrom 1998). And some oceanographers argued that P

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should be the master control on primary production in estuaries, as it has been viewed to be in oceanic waters since first proposed by Redfield (Smith 1984; Tyrrel 1999). The debate over the relative importance of N and P as drivers of coastal eutrophication has continued over much of the last two decades and has slowed progress toward effort to control N pollution (NRC 2000). Criteria for P control in both freshwaters and estuaries were proposed in the 1960s in the United States (Federal Water Pollution Control Administration 1968), yet the federal government only began the process to develop criteria for N regulation in estuaries in the past few years (EPA 2001), and to date, no criteria have actually been proposed. In Europe, N pollution as a contributor to coastal eutrophication was generally ignored by the governments of most European nations until well into the 1990s (Chave 2001; Howarth et al. in press).

In 1971, the American Society of Limnology and Oceanography (ASLO) held the "Symposium on Nutrients and Eutrophication: The Limiting-Nutrient Controversy," which led to the first-ever special issue of *Limnology and Oceanography*, "Nutrients and Eutrophication" (Likens, editor, 1972). By then, eutrophication was an obvious and growing problem in many aquatic ecosystems, and one that had grabbed the public's attention and spawned significant scientific debate with regard to its cause. Those who are aware of the controversy over N versus P as a primary control on eutrophication in estuaries may be surprised to find this topic virtually absent from the 1972 volume. The "limiting-nutrient controversy" discussed there focused on whether P or carbon (C) is more important in eutrophication. In the words of Likens (1972), "this symposium represents an attempt by the American Society of Limnology and Oceanography to provide a statement on the importance of various regulating or 'limiting' nutrients in the eutrophication of aquatic ecosystems. The subject is extremely timely because of the current so-called carbon-phosphorus controversy, and is loaded with political and economic overtones."

In this article, we briefly review the changing views on the role of N as the limiting nutrient for eutrophication in coastal marine ecosystems since the time of the 1971 symposium. We especially highlight the new types of evidence that came to the forefront in the 1980s and 1990s and have now led to the general consensus on the role of N in coastal eutrophication, and we briefly discuss some of the complexity on N and the related management of coastal eutrophication that has increasingly come to light in very recent years.

The 1970s: The development of separate lake and estuarine sciences of eutrophication

The 1971 ASLO symposium on the limiting-nutrient controversy was strongly focused on freshwater systems and on P versus C control of eutrophication. Only 3 of 19 papers in the 1972 special issue of *Limnology and Oceanography* from this symposium addressed estuarine or marine systems or organisms (Fuhs et al. 1972; Jaworski et al. 1972; Pomeroy et al. 1972). One of these studies (Pomeroy et al. 1972) concentrated almost exclusively on P biogeochemistry in es-

tuaries and barely mentioned N as a possible regulator of eutrophication. The Fuhs et al. (1972) study focused on P regulation of growth rates of pure cultures of a marine diatom, again without mentioning the role of N with regard to production of marine organisms. The Jaworski et al. (1972) manuscript stands out in that it is the only paper in that special issue that suggests that N may be the element that most limits primary productivity in an estuary, in this case the Potomac River estuary. Jaworski et al. (1972) also hinted that the controls on eutrophication in estuaries may differ from those in lakes. While some papers dating from much earlier in the 20th century concluded that primary production and eutrophication in marine ecosystems are limited by N (Johnston 1908; Ryther 1954), the Jaworski et al. (1972) paper offers, as far as we know, the first suggestion made in the peer-reviewed literature that the controls in lakes and estuaries differ. Only one other paper in the 1972 special issue suggests that N may ever limit production in an aquatic ecosystem, and it argued for co-limitation of P and N in an oligotrophic lake (Fuhs et al. 1972). Curiously, no papers in the special issue reference the work of Ryther (1954) and Ryther and Dunstan (1971) on N control of eutrophication in coastal marine ecosystems.

As noted by Schindler (1981), during the decade following the 1971 ASLO symposium, there was "little interaction between scientists studying the eutrophication of freshwaters and those studying the eutrophication of estuaries." The community of freshwater scientists continued to focus on P and by the middle to the end of the 1970s had developed a strong consensus that P caused eutrophication in lakes. The whole-lake experiments of Schindler and colleagues were instrumental in developing this consensus (Schindler 1974, 1977; Schindler et al. 1978), as were the cross-lake comparative P-loading models of Vollenweider (1975, 1976). This body of science led rather quickly toward governmental actions to better control P inputs to natural waters (Smith et al. 1999).

Interestingly, the community of freshwater scientists during the 1970s tended to adopt the ocean-scale view of Redfield (1958), which holds that P is the master element controlling productivity. In this view, any deficits in the N requirement of phytoplankton relative to their P requirements are made up through N fixation by planktonic cyanobacteria. The whole-lake experiments at the Experimental Lakes Area (ELA) clearly showed such a response, and significant planktonic N fixation occurred only in lakes in which the N:P ratio of nutrient inputs was below the Redfield ratio of 16:1 (Schindler 1977; Flett et al. 1980). In the Lake 227 experiments, this N fixation did indeed alleviate short-term N shortages and contributed to overall P limitation of net primary productivity in the lake. In support of this view, Smith (1983), in a comparative study of many lakes, found N-fixing species of cyanobacteria to be abundant in the plankton only in lakes in which the N:P ratios were fairly low.

The community of freshwater scientists also developed skepticism about the use of short-term bioassays to determine the causes of eutrophication (*see*, for example, Hecky and Kilham 1988). Many bioassay studies had suggested that C might regulate eutrophication in lakes, fueling much of

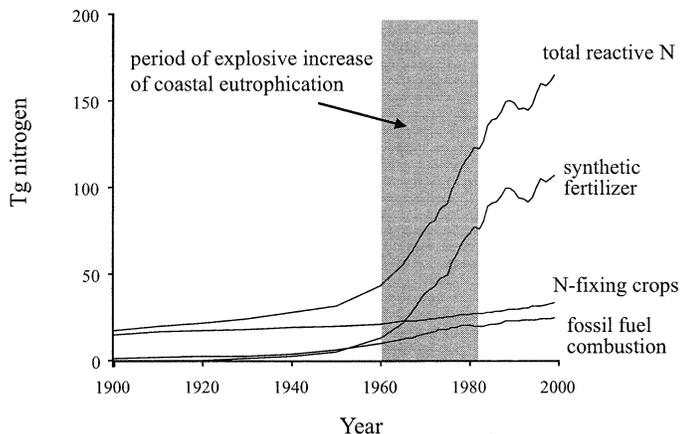


Fig. 1. Globally, the rate of creation of reactive nitrogen from human activity increased markedly over the past 50 yr, due largely to the manufacturing of synthetic fertilizer but also to combustion of fossil fuels. The global explosion of coastal eutrophication is coincident with this increase in reactive nitrogen. Figure modified from Boesch (2002). Data on global trends in nitrogen creation are from Galloway et al. (2004).

the P versus C limiting-nutrient controversy of the early 1970s. This led Schindler (1981) to write: “Laboratory studies to detect the ‘limiting nutrient’ have been shown to be of little use in developing nutrient strategies for lakes. Such studies conducted in Lake 227 showed that the lake was chronically carbon-limited throughout the summer months, for a period of several years. Yet years of observation have shown that all during that period the lake was in the process of slowly overcoming its carbon deficit by drawing and storing atmospheric CO₂, until finally, in 1975, after six years of fertilization, phytoplankton were no longer carbon limited. It is, of course, futile to tailor costly management strategies to the relatively short period of non-equilibrium when nutrient incomes to a water body are changed, when what is desired are control measures which will be effective for long periods.” This comment highlights the importance of time scale. In addition, bioassays in the lab are more likely to induce CO₂ limitation than would be the case in natural ecosystems, since in nature, CO₂ is more quickly replenished by diffusion from the atmosphere and from sediment respiration (Howarth 1988; NRC 1993). Other problems with the interpretation of bottle-assay experiments are reviewed by Hecky and Kilham (1988) and by Howarth and Michaels (2000).

The 1970s saw an explosive increase in coastal eutrophication in many parts of the world that was correlated with an accelerating rate of creation of reactive N from human activity (Fig. 1; Boesch 2002). During this decade, estuarine scientists began to concentrate on N as the cause of eutrophication in the ecosystems they studied. Bioassay studies in the coastal waters of New York (Ryther and Dunstan 1971), Cape Cod (Vince and Valiela 1973), Narragansett Bay (Smayda 1974), and the Oresund Straits between the Baltic and North Seas (Granéli 1978) indicated that N was much more important than P in regulating primary production. In 1979, the International Symposium on the Effects of Nutrient Enrichment in Estuaries was held in Virginia. In the 33

papers published from this symposium (Neilson and Cronin 1981), the overwhelming focus was on N, although many of the papers deal with P and silica (Si) as well. This community of scientists was clearly convinced that N was at least part of the story—if not the main story—behind eutrophication in coastal marine ecosystems. And interestingly, the proceedings of this symposium indicate a movement toward governmental regulation of N, in addition to P, in some localities—notably the Potomac River estuary (Bellanca 1981). However, as noted by Schindler (1981), this accumulating body of estuarine science had little influence in the community of freshwater scientists. This information also had little influence on the community of water-quality managers in either the United States or Europe, and there was no general trend toward developing N criteria and standards to control eutrophication in estuaries and coastal waters (NRC 1993, 2000; Chave 2001).

The 1980s: A continued emphasis on nitrogen in temperate estuaries and an eventual start toward freshwater-estuarine-marine comparisons

During the 1980s, research on nutrient dynamics and eutrophication in temperate-zone estuaries continued to focus on N, and most researchers who studied these systems continued to believe that eutrophication was controlled by N (NRC 1993, 2000). In part, this was based on bioassay work, such as the studies discussed above. Similar bioassay studies proliferated in the 1980s (D’Elia et al. 1986), with many but not all suggesting N limitation (*see* review by Howarth 1988). Until late in the 1980s, there is little evidence in the published literature that most coastal marine scientists were aware of the skepticism that their freshwater colleagues had toward the bioassay methodology (but *see* Smith 1984). One could argue that one of the fundamental criticisms of the bioassay approach—the induced and short-term CO₂ limitation in bottles (Schindler 1981)—would be less of a problem in coastal marine studies than in soft-water lakes because of the much greater concentrations of dissolved inorganic C in seawater (Howarth 1988). Nonetheless, there are a variety of other concerns with bioassay experiments, such as reduced turbulence, toxic effects from metals leaching from glass, unknown effects on the grazing community, and the rather short-time frame available for responses to occur (Howarth and Michaels 2000).

Other types of evidence used to infer N limitation of primary production in temperate estuaries were presented in the early 1980s. These included cross-system regressions showing primary production and chlorophyll to be correlated with N inputs (Boynton et al. 1982; Nixon and Pilson 1983), an approach similar to the P-loading curves of Vollenweider (1975, 1976) for freshwater systems. However, these early estuarine relationships were based on far fewer study systems than were used in the Vollenweider (1975, 1976) analyses and are therefore less convincing as a generality. Boynton et al. (1982) also inferred N limitation in many estuaries from the generally low ratios of dissolved inorganic N to P that characterize many of these systems during the time of peak primary production (Boynton et al. 1982). However,

this approach too can be criticized, since standing-stock concentrations of dissolved inorganic nutrients may not always accurately reflect their relative biological availabilities (Howarth 1988; Howarth and Marino 1990; Dodds 2003).

While coastal scientists studying temperate-zone estuaries during the 1980s usually concentrated on N as the regulator of primary production, those studying tropical marine coastal systems often concentrated on P. Net primary production in many shallow tropical systems with carbonate sands—at least in areas with little human influence—is limited by P (Smith and Atkinson 1984; Short et al. 1990), probably both because of high adsorption of P onto the sediments (Short et al. 1990; Howarth et al. 1995) and high rates of benthic N fixation by cyanobacterial mats, epiphytes, and symbionts (Smith 1984; Howarth et al. 1988*b*). Note that these tropical carbonate systems may become N limited as anthropogenic nutrient enrichment accelerates and rates of P sorption decrease (McGlathery et al. 1994; Howarth et al. 1995; Jensen et al. 1998). From his studies of N and P dynamics in low-nutrient tropical lagoons, Smith (1984) was impressed at how P acted as the master variable, with N being responsive over time, just as was the case in the ocean system described by Redfield (1958) and the lake experiments described by Schindler (1977). Smith (1984) generalized that P rather than N was the element likely to be limiting to production in most coastal marine ecosystems, including deeper, temperate systems and those that are nutrient enriched.

The 1980s saw some major synthetic work on N in marine ecosystems (*see* Carpenter and Capone 1983 for example) and began a greater move toward cross-fertilization of ideas and comparison of lakes and coastal marine ecosystems, as exemplified by another special issue of *Limnology and Oceanography*, “Comparative Ecology of Freshwater and Marine Ecosystems” (Nixon, editor, 1988). Several papers in that volume focused on differences in N and P cycling between lakes and estuaries, with a view toward how controls on production and eutrophication might vary (Froelich 1988; Howarth et al. 1988*a,b*; Seitzinger 1988). On the other hand, Hecky and Kilham (1988) focused on the evidence behind determinations of nutrient limitation in lakes and estuaries and concluded that: “A review of the experimental and observational data used to infer P or N limitation of phytoplankton growth indicates that P limitation in freshwater environments can be demonstrated rigorously at several hierarchical levels of system complexity, from algal cultures to whole lakes. A similarly rigorous demonstration of N limitation has not been achieved for marine waters. Therefore, we conclude that the extent and severity of N limitation in the marine environment remains an open question.” In a 1988 review, one of us (Howarth 1988) concluded that the preponderance of evidence did indeed suggest N regulation of primary production and eutrophication in many temperate-zone estuaries, but we also noted the need for whole-ecosystem experiments in estuaries of the sort done in freshwater lakes as a way to further the dialogue and better prove the importance of N as a controlling nutrient in temperate estuarine systems.

During the 1980s, the governmental regulation of P inputs to natural waters that began in earnest in the 1970s was proving its value, and the problems associated with eutro-

plication in freshwater lakes began to be reversed (Carpenter et al. 1998*a,b*). On the other hand, there was a continued explosion both in the United States and globally of problems associated with coastal eutrophication (Boesch 2002). In some locations, there was recognition by decision makers that N control might be necessary to address coastal eutrophication, notably in Tampa Bay and in the Chesapeake Bay in the United States (NRC 2000). However, this did not occur without significant political resistance. One state official in Maryland strongly complained to the administration of the University of Maryland about a faculty member there who was publicly pushing for regulation of N inputs to Chesapeake Bay based on the results of his research and that of colleagues (D. Boesch pers. comm.). In Europe, there was as yet no focus on coastal eutrophication, with the exception of a 1985 conference of countries bordering the North Sea, which called for a reduction of all pollutants (including P and N) to the North Sea by 1995 (Chave 2001). Overall, the research on N limitation of eutrophication in estuaries had little or no influence on the development of water-quality management at national scales throughout the 1980s, and in fact well into the 1990s (NRC 1993, 2000). National nutrient management strategies continued to focus on P, based on lake eutrophication studies and management strategies.

The 1990s: The development of a consensus for nitrogen control of coastal eutrophication—whole-system evidence

Throughout the 1990s, there was a growing trend toward a scientific consensus for the need to control N inputs to coastal marine ecosystems (Nixon 1995). This is clearly illustrated in a series of “white-paper” reports from the Ecological Society of America (Vitousek et al. 1997; Carpenter et al. 1998*a*; Howarth et al. 2000*b*) and reports from committees of the U.S. National Academy of Sciences (NRC 1993, 2000). This work slowly has had an influence on governments. By 2001 in the United States, the U.S. Environmental Protection Agency clearly recognized the need for N control and published the “Nutrient Criteria Technical Guidance Manual for Estuarine and Coastal Marine Waters” as a step toward meeting this need (EPA 2001). In the 1990s in Europe, the “Urban Wastewater Treatment Directive” called for reductions in loadings of both N and P from urban wastewater streams into sensitive coastal waters, and the “Nitrate Directive” called for better management of N from agricultural sources, although the driving interest behind this directive was nitrate in drinking water rather than coastal eutrophication (Chave 2001). In 2001 the European Union adopted a watershed-based approach (“The Water Framework Directive”) that is intended to replace these earlier directives and substitutes a watershed-based approach for managing both N and P in groundwater and freshwaters as well as coastal waters (Chave 2001; Howarth et al. in press). Smith et al. (1999) highlight several recent efforts to develop controls on N inputs to coastal waters in Europe.

The EPA (2001) “Nutrient Criteria Technical Guidance Manual for Estuarine and Coastal Marine Waters” relies heavily on the content of the NRC (2000) report for its jus-

tification. In the NRC (2000) report, the Committee on Causes and Management of Coastal Eutrophication stated that an increasing body of science in the 1990s led to a definite conclusion that N was the prime culprit behind eutrophication in many and probably most estuaries in the temperate zone. This conclusion was based in part on a series of whole-system-scale experiments and observations reported during the 1990s as well as on an improved mechanistic understanding of how the controls on eutrophication may vary between lakes and estuaries (NRC 2000). Ecosystem-scale experiments were the galvanizing force that led to the clear conclusion that eutrophication in lakes is best managed through controlling P inputs (Schindler 1977; Hecky and Kilham 1988; NRC 1993), and these estuarine experiments demonstrated a striking contrast in the behavior of the ecosystems.

One of these experiments was conducted at the Marine Ecosystem Research Laboratory (MERL) on the shores of Narragansett Bay (Rhode Island). This mesocosm experiment was specifically designed to see if coastal systems respond to nutrient additions in the same manner as lakes (Oviatt et al. 1995). Large mesocosms containing water and sediment from Narragansett Bay were maintained for a period of 4 months; several previous studies in the MERL facility had demonstrated that these systems accurately mimic much of the ecological functioning of Narragansett Bay. In this experiment, mesocosms received either no nutrient enrichment (control) or were enriched with N, P, or both (Fig. 2). The nutrient loadings paralleled those used in the ELA experiments (Schindler 1974, 1977). In sharp contrast to those whole-lake experiments, the addition of N (either alone or with P) but not of P alone to the MERL coastal mesocosms caused large increases in both rates of primary production and concentrations of chlorophyll (Oviatt et al. 1995).

Another mesocosm experiment on the shores of Narragansett Bay evaluated the response of shallow temperate seagrass systems typical of the northeastern U.S. coast to enrichment with N, P, or N+P (Lin et al. 1995; Taylor et al. 1995a,b). This experiment also showed no effect of P alone on chlorophyll in the water column. Chlorophyll abundance was stimulated to some extent by N alone but even more so by N+P together (Taylor et al. 1995a,b). The stimulation of chlorophyll was accompanied by a decline in seagrass biomass and productivity (Lin et al. 1995; Taylor et al. 1995a).

A third whole-ecosystem estuarine study followed the effects of experimental alteration of nutrient releases from a sewage treatment plant into Himmerfjarden, an estuary south of Stockholm, Sweden, on the Baltic Sea. The response of the estuary to nutrient inputs from sewage (the primary input to this system) was studied from 1976 to 1993 (Elmgren and Larsson 1997). For the first 12 yr, N loads gradually increased as population grew in the sewered district, while for the first seven of these years (until 1983), P loads were decreased as P-reduction technology was brought on line. For a 1-yr period beginning in the fall of 1983, P additions were then greatly increased (by stopping the P removal during sewage treatment). Subsequently, P removal was again used, but N inputs were increased by 40% as a result of an increase in population served by this particular sewage treatment plant. Finally, N-removal technology was gradually intro-

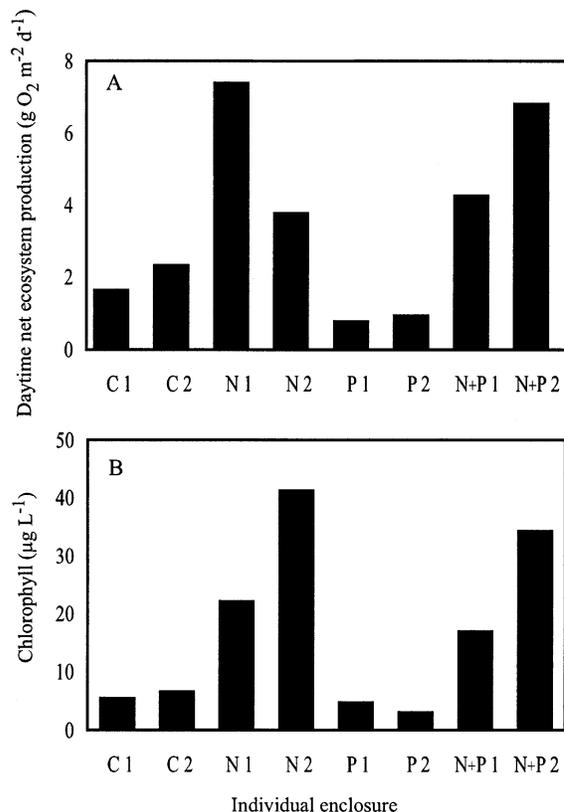


Fig. 2. Response of estuarine mesocosms at the MERL facility in Rhode Island to nitrogen and phosphorus additions. C1 and C2 were replicate control systems; N1 and N2 were replicate systems fertilized with nitrogen; P1 and P2 were replicate systems fertilized with phosphorus; and N+P1 and N+P2 were replicate systems fertilized with both nitrogen and phosphorus. Nitrogen, either alone or in combination with phosphorus additions, stimulated both (A) production and (B) chlorophyll levels. Phosphorus alone stimulated neither production nor chlorophyll levels and may actually have somewhat depressed these. Data show average responses over the course of the experiment. Reprinted from Oviatt et al. (1995).

duced to the sewage treatment plant between 1988 and 1993, reducing the N load to the value originally seen in 1976. Throughout the 17 yr of observation, the concentration of total N tended to reflect the nitrogen input from the sewage treatment plant (Elmgren and Larsson 1997), and both abundance of phytoplankton and water clarity were clearly related to this. Total P concentrations varied independently of total N over time in Himmerfjarden, and total P was a poor predictor of phytoplankton abundance. These data provide strong evidence that N was the primary element controlling eutrophication in this estuary. During the year that P loadings were experimentally increased, there was no effect on primary production; however, there was an unusually large phytoplankton bloom the following spring, probably due both to some residual high levels of P and to an unusually high input of N from spring floods (Elmgren and Larsson 1997).

A fourth whole-ecosystem study explored long-term changes in Laholm Bay, an estuary on the southwestern coast of Sweden (Fig. 3). Early signs of eutrophication ap-

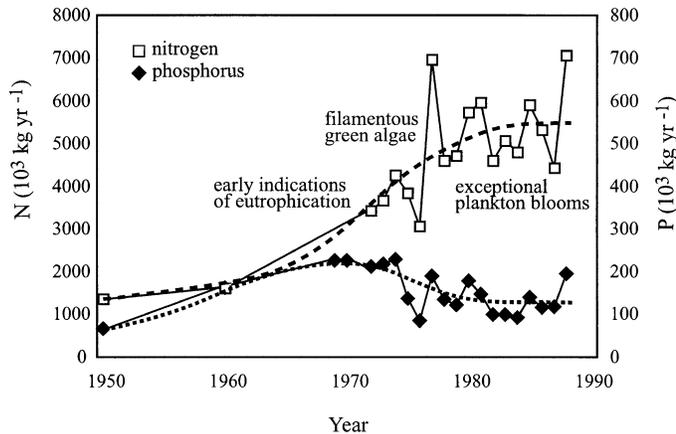


Fig. 3. Time trend of nutrient enrichment into Laholm Bay in southwestern Sweden from 1950 until 1988. Phosphorus loads decreased after 1970 in response to management for eutrophication, while nitrogen loads, which were unregulated, continued to increase. Early evidence of eutrophication in the Bay appeared in the 1960s. Eutrophication grew steadily worse over the time period in which phosphorus loads decreased and nitrogen loads increased. Reprinted from Rosenberg et al. (1990).

peared there in the 1950s and 1960s and steadily increased over time (Rosenberg et al. 1990). The earliest reported signs of eutrophication were changes in the community composition of macroalgal species, and over time, filamentous algae typical of eutrophic conditions have become more prevalent. Harmful algal blooms in the water column have become much more common, particularly in the 1980s (Rosenberg et al. 1990). During the early stages of eutrophication in Laholm Bay, inputs of both P and N to the estuary were increasing. However, from the late 1960s through the 1980s, P inputs decreased by a factor of almost two as a result of stringent management. N inputs, which were not managed, continued to rise and almost doubled over time (Rosenberg et al. 1990). During this same period, plankton blooms continued, clearly indicating that N and not P controlled the Laholm Bay eutrophication.

These whole-system-scale experiments and observations demonstrate that N was the primary control on eutrophication in specific temperate zone estuaries (Narragansett Bay, Himmerfjorden, and Laholm Bay). Importantly, however, the finding in each of these three systems is consistent with conclusions drawn from short-term bioassay studies and from inference of the relative N and P availability based on ratios of dissolved inorganic N:P in these ecosystems (Granéli et al. 1990; Oviatt et al. 1995; Elmgren and Larsson 1997). These ecosystem-scale studies therefore add credence to the application of bioassay data and inorganic nutrient data in assessing whether N or P is more limiting in estuaries (NRC 2000). The large preponderance of bioassay data in estuaries and coastal marine systems indicates N limitation (Howarth 1988), as does the generally low inorganic N:P ratio found in many estuaries at the time of peak primary production. Thus, taken together, results from the whole-system-scale studies and bioassay data and N:P ratio data from many sites led to the conclusion that N availability is the primary

regulator of eutrophication in most temperate coastal systems (NRC 2000).

Another estuarine “field” experiment that supports conclusions drawn by the NRC (2000) began in the early 1960s, this one the result of P-mining operations in the watershed of the Pamlico River estuary in North Carolina. The leakage of large amounts of P into the estuary over a period of many years had no demonstrable effect on water quality, very strongly indicating that the system was not P limited (Stanley 1993). This important result has never been published in the peer-reviewed literature, as far as we know, and unfortunately has attracted very little notice.

Understanding the mechanistic differences in nutrient limitation between lakes and estuaries

In addition to the whole-system-scale evidence for the importance of N that emerged during the 1990s, the NRC (2000) report summarized the growing body of evidence for a plausible mechanistic difference between the functioning of lakes and estuaries with regard to N versus P control. Whether primary production by phytoplankton is N or P limited is broadly framed in terms of the relative availabilities of N and P in the water compared to the stoichiometric need for average cell growth (the so-called Redfield ratio of 16:1). Nutrient availability in turn is determined by the ratio of external N:P in inputs to the ecosystem, preferential storage, recycling, or loss of N or P in the ecosystem, and the amount of biological N fixation (Howarth 1988; Vitousek and Howarth 1991). One critical difference between lakes and estuaries regards the importance of N fixation. In mesotrophic and eutrophic lakes, shortages of N relative to P tend to be made up through planktonic N fixation by cyanobacteria, and this is one of the mechanisms that lead to P regulation of primary production and eutrophication (Schindler 1977; Flett et al. 1980). This N fixation response generally does not occur in estuaries with salinities >10 to 12, even when they are strongly N limited (McCarthy 1980; Howarth et al. 1988b; Paerl 1990). Controls on planktonic N fixation in estuaries and evidence for mechanistic differences with freshwaters are discussed in detail in the next section.

A second major difference between freshwater and coastal marine systems regards the nutrient inputs. Lakes receive nutrient inputs from upstream terrestrial ecosystems and from the atmosphere, while estuaries and coastal marine systems receive nutrients from these sources as well as from neighboring oceanic water masses. For estuaries such as those along the northeastern coast of the United States, the oceanic inputs tend to have an N:P ratio that is well below the Redfield ratio as a result of the denitrification on the continental shelves (Nixon et al. 1995, 1996). Thus, given similar nutrient inputs from land, estuaries are more likely to have total nutrient loads with a lower N:P ratio than lakes, more strongly favoring N limitation (NRC 2000). In addition, many urban centers are near the coast and thus often have a greater influence on nutrient loads to estuaries and coastal systems than to lakes. The N:P ratio in nutrient loads from urban areas tends to be quite low, further favoring

N limitation in the coastal systems that receive such loads (Billen et al. 1991).

A third major difference between lakes and estuaries favoring P limitation in freshwaters and N limitation in saline waters is the greater relative availability of P in estuaries. One mechanism leading to the greater availability of P in estuaries is the desorption of P from suspended matter born down rivers as the salinity increases and other anions compete with phosphate for sorption sites (Froelich 1988; Howarth et al. 1995). Another mechanism is the tendency for greater storage of P in sediments and anoxic bottom waters in lakes than in estuaries (Caraco et al. 1989, 1990; Blomqvist et al. 2004). The tendency for lower storage of P in estuarine sediments relative to freshwater sediments is probably the result of higher sulfate concentrations, leading to higher sulfate reduction rates and more sequestration of iron by sulfides. Iron sulfides are less adsorptive of phosphate than are other iron minerals (Krom and Berner 1980). Although estuarine sediments in general are less likely to adsorb and immobilize P than are lake sediments (Caraco et al. 1990; Blomqvist et al. 2004), the importance of this process is variable among estuaries (Sundby et al. 1992; Howarth et al. 1995). At one extreme, little or no P is adsorbed by the sediments of Narragansett Bay, and virtually all of the phosphate produced during decomposition in the sediments is released back to the water column (Nixon et al. 1980). This, in combination with N lost through denitrification, is a major reason that primary production in Narragansett Bay is predominantly N limited (Nixon et al. 1980; Howarth 1988). On the other hand, the P released from sediments in Chesapeake Bay is less than the amount released during decomposition (Boynton and Kemp 1985), and there is evidence of P adsorption and storage in surface sediments in several other estuaries (van Raaphorst et al. 1988; Koop et al. 1990; Sundby et al. 1992). Many of the differences in P uptake and release from sediments in both estuaries and lakes may be explained in part by the extent of eutrophication in those systems. There is often a positive feedback of P release with increased eutrophication, as more organic carbon leads to more reducing sediments (Schindler 1981; Howarth et al. 1995). Thus, the potential for N limitation in estuaries likely increases with increased nutrient loading.

Is there also a difference in the importance of denitrification between lakes and estuaries? Denitrification is often a major sink for N, and it tends to drive systems toward N limitation unless counterbalanced by other processes such as P adsorption and storage in sediments (Howarth 1988; Seitzinger 1988; Nixon et al. 1996). The overall magnitude of denitrification tends to be greater in estuaries than in freshwater ecosystems, but this may simply be a result of greater N fluxes through estuaries (Seitzinger 1988). When expressed as a percentage of the nitrogen input to the system lost through denitrification, there appears to be relatively little difference between estuaries and freshwater ecosystems (Nixon et al. 1996). That is, available evidence indicates that denitrification tends to drive both coastal marine and freshwater ecosystems toward N limitation, with no greater tendency in estuaries (NRC 2000). In fact, the tendency toward nitrogen limitation—based on this process alone—might be greater in lakes, since lakes generally have a longer water

residence time, and the percent N loss through denitrification is greater in aquatic ecosystems having longer water residence times (Howarth et al. 1996; Nixon et al. 1996; Seitzinger et al. 2002).

Planktonic nitrogen fixation:

The unique biogeochemical space of estuaries

As noted above, a long tradition of thought by oceanographic scientists has held that P is the long-term regulator of primary production in the oceans as a whole (Redfield 1958; Broecker 1974). In this view, N limitation can occur in oceanic surface waters, but this is a transient effect that is made up for by N fixation by planktonic cyanobacteria in the surface waters over geological time scales. Karl et al. (2002) have suggested that there might be short-term variation in the nature of this response, leading to changes in the relative importance of N and P as limiting nutrients to production in oceanic waters on the time scale of years to decades. Nonetheless, the paradigm that P is the master variable controlling ocean-scale production over long time periods is still widely accepted. Schindler (1977) demonstrated the applicability of the concept to lakes, showing that planktonic N fixation can alleviate N shortages and maintain P limitation of primary production and eutrophication over much shorter time scales. Saline estuaries generally do not show this behavior, and cyanobacteria capable of N fixation are either present in very small numbers or are absent from the plankton of the vast majority of mesohaline and saline estuaries with salinities of >10 to 12 (McCarthy 1980; Howarth et al. 1988*b*; Paerl 1990). This lack of planktonic N fixation in even strongly N-limited estuaries helps maintain a deficiency in N availability relative to P (Howarth 1988; Vitousek and Howarth 1991; NRC 2000).

Much research has been directed at the question of why N fixation by planktonic organisms might differ between lakes and estuaries, with much of this focused on single-factor physical or biogeochemical controls, such as short residence times, turbulence, salinity, or limitation by iron, molybdenum, or P (*see*, for example, Paerl 1985 and Howarth et al. 1988*a*). A growing consensus has developed, however, that N fixation in marine systems, including estuaries, coastal seas, and also oceanic waters, likely is regulated by complex interactions of chemical, biotic, and physical factors (Paerl and Zehr 2000; Karl et al. 2002; Marino et al. 2002). Similarly, the controls on N fixation in terrestrial ecosystems probably involve a complex set of interactions (Vitousek et al. 2002). With regard to estuaries and coastal seas, recent evidence indicates that the ecological interaction of two mechanisms—a combination of slow growth rates and grazing mortality by zooplankton and benthic animals—can preclude N-fixing heterocystic cyanobacteria populations from developing (Howarth et al. 1999; Marino et al. 2002, 2003). The slow growth rate of cyanobacteria in saline estuaries relative to those in freshwater can be caused by the higher sulfate concentrations found in seawater (Howarth et al. 1988*a*; Marino et al. 2003; Stal et al. 2003). In estuarine mesocosms experiments with saline water (>30), planktonic heterocystic cyanobacteria grew and fixed N when the abun-

dance of macro- and meso-zooplankton grazers was kept low, but numbers of cyanobacteria and the quantity of N fixed was far lower than in a freshwater experiment with similar controls on physical factors (turbulence and mixing depth, residence time) and nutrient availability (high P and low N:P; Marino et al. 2002, in press).

In coastal marine ecosystems where the salinity is low, N fixation by planktonic cyanobacteria is often observed, notably in the Baltic Sea and its tributaries (Wallstrom et al. 1992; Moisaner et al. 2003; Stal et al. 2003) but also the Peel-Harvey inlet in Australia (Huber 1986), the Neuse River estuary in North Carolina (Piehler et al. 2002), and several other coastal ponds and estuaries. For most of these ecosystems, planktonic N fixation is only observed when the salinity is <10 to 12, and generally much lower yet. As far as we are aware, N-fixing cyanobacteria have not been reported in any estuary or coastal sea where the salinity is persistently >10 to 12. One explanation for the occurrence of N-fixing cyanobacteria in the plankton of these low salinity systems (but not in most estuaries and coastal marine ecosystems at higher salinities) is that the sulfate concentrations are considerably lower. The lower sulfate is predicted to lead to growth rates by cyanobacteria that are high enough to escape the grazing control by zooplankton and lead to cyanobacterial populations and rates of N fixation that are similar to those observed in freshwaters (Howarth et al. 1999; Marino et al. 2002). In the Peel-Harvey estuary and a similarly shallow, highly nutrient-enriched estuary in Tasmania, blooms of N-fixing cyanobacteria begin during the wet season, when the estuaries are largely fresh or very low salinity and then persist as salinity gradually increases to levels well above 12 (Huber 1986; Lukatelich and McComb 1986; Jones et al. 1994). The low salinity (and consequently low sulfate concentrations in comparison to saline estuaries) alone may be sufficient explanation for the abundance of N-fixing, planktonic cyanobacteria in these estuaries. Further, the extreme state of eutrophication also may encourage planktonic N-fixing cyanobacteria, since water-column anoxia may increase trace metal availability (iron and molybdenum) as well as decrease grazer populations (Howarth et al. 1988a, 1999; Howarth and Marino 1998).

An interesting side note concerns the evidence for the abundance of heterocystic cyanobacteria in estuaries. The community of estuarine scientists has long perceived that planktonic, N-fixing cyanobacteria are scarce or absent from most saline estuaries (salinities >10 to 12). For example, McCarthy (1980) noted "the phytoplankton capable of N₂ fixation are relatively common in fresh water, scarce in the open sea and virtually unknown in estuaries." However, the scientific literature often does not adequately reflect negative findings, and in fact very few papers have reported the absence of planktonic, heterocystic cyanobacteria and the absence of measurable N fixation by plankton in saline estuaries (see reviews by Howarth et al. 1988a,b for the status as of the 1980s; this remains so today). Hence, most of the published studies on N fixation by plankton in estuaries and coastal seas focus on the relatively rare systems in which rates are measurable, such as the Peel-Harvey estuary or the low-salinity or freshwater portions of estuaries. Recent work has examined genetic evidence for N fixation in estuaries:

for example, looking for the presence of the *nifH* gene in water samples. While interesting, the presence of *nifH* does not necessarily indicate gene expression, and RNA provides a better determination of whether *nifH* is expressed under in situ conditions (Zehr et al. 2001). In fact, Piehler et al. (2002) found little correlation between *nifH* presence and nitrogenase activity in the oligohaline Neuse River estuary. We are aware of no evidence to date for either the presence or expression of the *nifH* gene in saline estuaries (salinities >10 to 12).

While planktonic N fixation appears to be a rare process in saline estuaries and coastal seas, benthic cyanobacteria and cyanobacterial epiphytes on seagrasses often fix N at high rates and so can contribute significantly to the N inputs of systems in which sufficient light penetrates to the bottom (Howarth et al. 1988b; Joye and Paerl 1993, 1994; Newell et al. 2002). As discussed above, benthic N-fixation rates were high in the shallow tropical lagoons studied by Smith and Atkinson (1984), which undoubtedly contributed to the view of Smith (1984) that estuaries follow the Redfield paradigm of N fixation being responsive to N:P ratios, as also occurs in lakes. However, benthic N fixation by cyanobacteria can only occur in those ecosystems in which sufficient light reaches the benthos, which is typically not the case in deeper, turbid, nutrient-enriched estuaries.

Toward a more complex view of eutrophication in estuaries

A strong consensus has developed over the past 5 to 10 yr that solving the problem of eutrophication in estuaries requires controls on N inputs (Howarth et al. 2000b; NRC 2000; Boesch 2002). There has also been a growing tendency among estuarine and coastal scientists to view eutrophication in a more complex manner. Simple models such as those developed by Vollenweider (1975, 1976) for P in lakes have substantial explanatory power, and indeed for many marine ecosystems, there is a positive linear relationship between inorganic N loading and rates of primary production (Boynton et al. 1982; Nixon et al. 1996). However, there is also variation in the response of estuaries to N loading, with some estuaries being far more sensitive to eutrophication than others (NRC 2000). In part, this is due to the complex interaction of nutrient limitation and light limitation in estuaries (Cloern 2001), as well as to the influence of residence time on community structure and ecological interactions, particularly in estuaries with relatively short residence times (Howarth et al. 2000a). These interactions are still relatively poorly understood, and an improved understanding of the factors that determine the sensitivity of estuaries to nutrients may eventually lead to better management of coastal nutrient pollution (NRC 2000; Howarth et al. 2003).

Further, a linear response of eutrophication to N loading does not appear to apply in shallow estuarine ecosystems, which are dominated by benthic primary producers such as seagrasses or algal mats. In such systems, primary production is often quite high even when external N inputs are low, and in contrast to deeper systems, the effect of N loading on primary production is quite nonlinear (Nixon et al. 2001).

The high productivity in these systems at low levels of external N loading is likely supported by high rates of N fixation by benthic and epiphytic cyanobacteria. As N loading increases, this N fixation may be gradually suppressed, so that the total inputs of N to the shallow systems (from N loading and N fixation) remain rather constant over a range of external N loads and then increase disproportionately.

Nitrogen, while clearly very significant, is not the only element of concern for coastal systems, even for those in the temperate zone, and in some estuaries, P is probably limiting (Howarth 1988). For instance, P may be limiting in the Apalachicola estuary on the Gulf coast of Florida and in several estuaries on the coast of The Netherlands in the North Sea (Myers and Iverson 1981; Postma 1985; Brockman et al. 1990). In the case of the North Sea estuaries, P limitation is probably the result of extremely high N inputs combined with fairly stringent control of P inputs (Howarth et al. 1995, 1996). In the case of the Apalachicola, P limitation results from a relatively high ratio of N:P in nutrient inputs, although in this case, the high ratio may reflect the relatively small amount of human disturbance in the watershed and the relatively low nutrient inputs overall (Howarth 1988; Billen et al. 1991). P also limits primary productivity in the eastern Mediterranean Sea, probably because a high input of iron in dust from the atmosphere results in precipitation of P from the water column (Krom et al. 1991).

Nutrient limitation of primary production may switch seasonally between N and P in some major estuaries, such as Chesapeake Bay (Malone et al. 1996), and in portions of the Gulf of Mexico, including the "dead zone" (Rabalais et al. 2002a,b). In these systems, N is probably the nutrient responsible for the major effects of eutrophication. The production of most of the biomass that sinks into bottom waters and leads to low-oxygen events is more likely to be controlled by N than by P; when primary production is P limited in these systems, relatively little of the production tends to sink out of the water column (Gilbert et al. 1995; Malone et al. 1996; Rabalais et al. 2002a,b). On the other hand, Conley (2000) has suggested that reducing P inputs to heavily nutrient-loaded coastal systems may further help alleviate problems from oxygen depletion, particularly in deep estuaries. Cugier et al. (2005) note that reducing P to the Seine River in France may be the most effective measure for reducing eutrophication there, but that reducing P without also reducing N inputs might simply result in longer-range transport of N to surrounding coastal areas. They conclude that control of both N and P is important.

The effects of eutrophication are not simply increased production and lower oxygen levels, and eutrophication frequently leads to changes in ecological structure in benthic fauna and flora and in the phytoplankton composition (NRC 2000). Some of these changes are driven by P availability, as the relative availabilities of N and P can select for domination by different primary producers. For example, high levels of P can encourage domination by some undesirable macro-algal species in the benthos (Conley 2000). Other changes are driven by Si availability, since Si is required by diatoms but not other types of phytoplankton. Eutrophication tends to lower the availability of Si, both absolutely and relative to N and P, resulting in a loss of diatoms from the

community (Conley et al. 1993; Justic et al. 1995; Cugier et al. 2005). The loss of diatoms has a profound effect on energy flow through the food web, as diatoms are relatively high in food quality (Turner et al. 1998). This shift may also favor harmful algal blooms (Smayda 1997; Conley 2000; Cloern 2001). Once Si is depleted, high levels of inorganic P may further promote harmful algal blooms, including *Phaeocystis* and many species of dinoflagellates, particularly when the N:P ratio is low (Glibert and Pitcher 2001; Cugier et al. 2005). Decreasing Si availability and the consequent lower abundances of diatoms also lower the sedimentation of organic matter into bottom waters, and thereby have a partially mitigating influence on low-oxygen events associated with eutrophication. In many coastal systems there may, however, still be sufficient Si to fuel diatom blooms during the critical spring bloom period when the majority of sedimentation often occurs (Conley et al. 1993; Turner et al. 1998). Further, eutrophication can lead to other complex shifts in trophic structure that might either increase or decrease the sedimentation of organic carbon (Turner et al. 1998).

Nitrogen controls to coastal waters are an essential part of reducing coastal eutrophication, since N is the element most limiting to production in most coastal marine ecosystems in the temperate zone, and over the past decade there has been increasing interest by governments in both the United States and Europe in controlling N. However, for the reasons discussed above, it is important that N-control strategies for coastal waters go hand in hand with P control, as called for in the EU Water Framework Directive (Chave 2001) as well as planning efforts for many coastal systems in the United States (NRC 2000; Boesch 2002). Additionally, managing water quality in the freshwater ecosystems upstream of coastal systems can be beneficial to water quality in the coastal marine ecosystems. For example, eutrophication in upstream freshwater ecosystems can reduce the Si flow down rivers due to sedimentation of diatoms as the freshwaters become more productive (Humborg et al. 2000). Controlling P inputs to the watershed as a whole, therefore, can help maintain a greater supply of Si to coastal waters, a highly desirable result (Cugier et al. 2005). The optimal management of coastal nutrient pollution should consider these complex interactions.

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