

Measured and modelled effects of temperature, dissolved oxygen and nutrient concentration on sediment-water nutrient exchange

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Abstract

Empirical models of sediment-water fluxes of NH_4^+ , NO_3^- and PO_4^{3-} were formed based on published reports. The models were revised and parameters evaluated based on laboratory incubations of sediments collected from Gunston Cove, VA. Observed fluxes ranged from -18 (sediment uptake) to 276 (sediment release) $\text{mg NH}_4^+ \text{m}^{-2} \text{day}^{-1}$, -17 to -509 $\text{mg NO}_3^- \text{m}^{-2} \text{day}^{-1}$, and -16.4 to 8.9 $\text{mg PO}_4^{3-} \text{m}^{-2} \text{day}^{-1}$. The model and observations indicated release of NH_4^+ was enhanced by high temperature and by low DO. Uptake of NO_3^- was enhanced primarily by high NO_3^- concentration and to a lesser extent by high temperature and by low DO. Direction of PO_4^{3-} flux depended on concentration in the water. Release was enhanced by low DO. No effect of temperature on PO_4^{3-} flux was observed.

Introduction

Estimates of the magnitude and direction of sediment-water nutrient exchanges are crucial to investigations of eutrophication in tidal waters. Sediment nutrient fluxes cannot be measured everywhere at all times, however. One approach to describe sediment nutrient fluxes in the absence of observations is to employ empirical models that predict flux as a function of conditions in the water column. Model formulations and parameter values may be selected from the literature or derived from measures on the water body of interest. In this report, the applicability of empirical models derived from data found in the literature is tested against observations collected in a specific water body. Functional forms are pre-

sented, parameters are evaluated, and variables most influential in determining flux are identified.

The study system

Gunston Cove is located on the Virginia shore of the Potomac River, 26 km downstream of Washington, DC. The Cove consists of freshwater, is 1 to 2 m deep, and has a surface area of 5 km^2 . Tide range in the Cove is 60 cm and currents are less than 10 cm sec^{-1} . Freshwater flow is largely provided by a municipal treatment plant with a capacity of $1.6 \text{ m}^3 \text{ sec}^{-1}$. Waters of the Cove are eutrophic and are characterized, during summer, by total nitrogen $> 1 \text{ mg L}^{-1}$, total phosphorus $> 0.1 \text{ mg L}^{-1}$, dissolved oxygen $> 5 \text{ mg L}^{-1}$, and chlorophyll 'a' $> 60 \text{ } \mu\text{gm L}^{-1}$.

Sediment cores collected in the Cove exhibit three distinct layers: a light-brown, flocculent surface layer a few mm thick, a medium-brown layer extending 1 to 2 cm into the sediments, and a black layer extending to the bottom of the cores. The upper cm of the sediments are penetrated by worm tubes, and *Chironomid* larvae are present occasionally.

Methods

Intact sediment cores with overlying water were collected in transparent acrylic columns (60 cm × 10 cm). Sediments occupied half the length of the columns. Additional water was collected at the same time and filtered (1 μm) to remove most particulate matter. Cores were put on ice, transported to a laboratory, and then placed in a constant-temperature bath at the temperature specified for incubation. Water overlying the sediments was replaced with 2.3 L of filtered water. Each column was closed with an opaque, air-tight cap fitted with stirrer and dissolved oxygen (DO) probe. Stirring provided sufficient turbulence for the DO probe to read correctly without resuspending bottom sediments. The column cap and the sides of the constant-temperature bath prevented light from reaching the sediment surface, simulating the light attenuation of Gunston Cove.

DO was initially brought to the desired level by bubbling gaseous oxygen or nitrogen through the water. DO was controlled by a Commodore VIC-20 microprocessor fitted with an analog-digital conversion unit. The VIC monitored each of the DO probes. When respiration drew DO below a specified level, a computer-operated valve opened and gaseous oxygen bubbled through the water. When the desired DO was again attained, the valve closed. Undissolved oxygen was released through a one-way valve in the cap of the column. This system regulated DO within ± 0.25 mg L⁻¹. Measurement of nutrient fluxes commenced when the cores reached the desired DO and temperature. Approximately 12 hours elapsed between collection of the cores and

initiation of an incubation. Incubation lasted 24 hours during which five 125 mL water samples were collected at six-hour intervals. The volume withdrawn was replaced with DO and temperature-adjusted, filtered water.

Incubations were conducted at eighteen different DO-temperature combinations. DO was fixed at 0, 2, 5, or 8 mg L⁻¹ and temperature was varied from 10 to 30 °C in five-degree increments. Four cores were incubated at a single temperature each week. Two cores were maintained at 0 or 2 mg L⁻¹ DO and two cores were maintained at 5 or 8 mg L⁻¹ DO. Sediment nutrient fluxes were indicated by concentration changes in the overlying water. A single column filled with only water served as a blank.

Cores were collected from late July to early October 1984. Sediments were incubated at one DO-temperature combination and discarded. Nutrient concentration at the initiation of an incubation was approximately the concentration in the Cove at the time of sample collection (Table 1). Use of fresh cores in each incubation minimized the possibilities of core 'burnout' and bacterial colonization of column walls. Properties of the sediments may have varied throughout the study, however. Incubations were repeated at two DO-temperature combinations to examine the potential effect on nutrient fluxes of week-to-week variations in sediment properties.

Ammonium (NH₄⁺) was analyzed by an automated phenate method, nitrate + nitrite (NO₃⁻) by the cadmium reduction method, and phosphate (PO₄³⁻) by an ascorbic acid method (American Public Health Association, 1985). Parameters in the empirical models were evaluated through non-linear regression (Robinson, 1985). Variables were entered in regression singly, in order of importance as indicated by the statistic R². Variables retained in the models were significant at the one-percent level.

Empirical models

Sediment NH₄⁺ release generally increases as a function of temperature (Bowden, 1984; Hale,

1975; Boynton *et al.*, 1980). Evidence exists that NH_4^+ release increases at low DO concentrations (Fillos and Molof; 1972). These observations fit the empirical model

$$F = F_0 \theta^{T-20} e^{\alpha \text{DO}} \quad (1)$$

in which F is release rate ($\text{mg NH}_4^+ \text{ m}^{-2} \text{ day}^{-1}$), F_0 is release rate at 20°C , 0 mg L^{-1} DO, T is temperature ($^\circ \text{C}$), DO is dissolved oxygen (mg L^{-1}), θ is a constant that expresses enhancement of release by temperature, and α (L mg^{-1}) is a constant that expresses suppression of release by DO.

Table 1. Sediment-water NH_4^+ , NO_3^- , PO_4^{3-} fluxes.

	$^\circ \text{C}$	DO	0	2	5	8
NH_4^+ initial concentration	10			0.50		0.49
	15		0.94	0.76	0.86	0.69
	20		0.22	0.25	0.84	0.25
	25		1.24	0.78	1.21	0.18
	30		0.34	2.16	0.31	1.99
NH_4^+ flux	10			75		76
	15		78	46	61	72
	20		161	44	48	-18
	25		160	111	50	79
	30		134	276	180	98
NO_3^- initial concentration	10			1.53		1.37
	15		2.09	1.06	2.09	1.01
	20		1.33	1.35	1.43	1.39
	25		2.41	1.37	2.39	1.25
	30		2.82	2.76	1.81	2.81
NO_3^- flux	10			-86		-35
	15		-166	-91	-84	-17
	20		-173	-165	-70	-64
	25		-304	-62	-229	-64
	30		-509	-440	-191	-244
PO_4^{3-} initial concentration	10			**		0.066
	15		0.022	0.089	0.022	0.033
	20		0.039	0.060	0.065	0.123
	25		0.047	0.019	0.053	0.050
	30		0.019	0.029	0.037	0.036
PO_4^{3-} flux	10			-6.4		0.2
	15		2.1	-0.8	-1.9	-2.3
	20		6.5	8.9	-4.0	-16.4
	25		7.5	3.9	-1.6	-9.3
	30		6.4		-4.3	0.0

Flux in $\text{mg m}^{-2} \text{ day}^{-1}$. Concentration in mg L^{-1} . Negative fluxes are from water to sediment. (**) PO_4^{3-} concentration below detection limit.

Sediment NO_3^- flux is enhanced by high temperature (Jenkins & Kemp, 1984; Messer & Brezonik, 1984; Seitzinger *et al.*, 1984; Klapwijk & Snodgrass, 1982), by low DO (Van Kessel, 1977a), and by high NO_3^- concentration in the water (Messer & Brezonik, 1984; Nedwell, 1982; Van Kessel, 1977a). These observations fit the empirical model

$$F = a C_w \theta^{T-20} e^{\alpha \text{DO}} \quad (2)$$

in which 'a' (mm day^{-1}) is a constant that expresses enhancement of uptake due to concentration, C_w is NO_3^- concentration in water (mg L^{-1}) and the remaining parameters are as defined for NH_4^+ .

Sediment PO_4^{3-} release may be enhanced at low DO (Fillos & Molof, 1972; Fillos & Swanson, 1975; Bates & Neafus, 1980; Holdren & Armstrong, 1980; Nixon *et al.*, 1980; Mawson *et al.*, 1983) and at high temperature (Fillos and Swanson, 1975; Nixon *et al.*, 1980, Hale, 1975; Boynton *et al.*, 1980). Reports exist of an interaction between PO_4^{3-} flux and concentration in the water column (Pomeroy *et al.*, 1965; Twinch & Peters, 1984; Fillos and Biswas, 1976). These observations fit the empirical model

$$F = F_0 \theta^{T-20} e^{\alpha \text{DO}} - a C_w \quad (3)$$

in which parameters are analogous to those defined for NH_4^+ and NO_3^- .

Results

Fluxes are reported as the average flux in replicate sediment columns and are corrected for apparent fluxes in the blank (Table 1). The sediments released NH_4^+ in all but one case. Release was enhanced at high temperature and low DO. Movement of NO_3^- was from the water to the sediments in all observations. Uptake was enhanced by high temperature and by low DO. Phosphate moved in both directions across the sediment-water interface. Sediments tended to release PO_4^{3-} at low DO. No effect of temperature was apparent.

The experimental approach allowed for comparison of nutrient fluxes measured in replicate cores and in repeat incubations using sediments collected three to five weeks apart (Table 2). Agreement between replicate cores and between repeat incubations was best for NO_3^- . Agreement of NH_4^+ fluxes measured in replicate cores was comparable to NO_3^- , but NH_4^+ flux measures were the least repeatable. Measures of PO_4^{3-} flux in replicate cores were less precise than NH_4^+ flux measures but PO_4^{3-} flux measures repeated several weeks apart agreed more closely than repeated NH_4^+ measures.

Parameters in the empirical models are presented in Table 3. Regression analysis indicated temperature was the most significant determinant of NH_4^+ flux, accounting for 42% of the variability observed. DO accounted for 12% of the observed variability. Concentration of NO_3^- was the most significant determinant of sediment uptake, accounting for 52% of the variability observed. Temperature and DO accounted for 24% and 13% of the total variability, respectively.

Table 2. Precision and repeatability of flux determination (CV).

	NH_4^+	NO_3^-	PO_4^{3-}
Median CV of replicate cores	0.22	0.18	0.87
Range of CV of replicate cores	0.03 to 1.31	0.04 to 1.29	0.04 to 2.34
CV of two repeat incubations	0.62, 2.22	0.0, 0.64	0.39, 1.68

Table 3. Model parameter values.

	NH_4^+	NO_3^-	PO_4^{3-}
F_0	108		10.2
θ	1.070	1.055	
α	-1.066	-0.083	-0.184
a		110	134
Total R^2	0.54	0.89	0.75
	$0.005 < p < 0.01$	$p < 0.001$	$p < 0.001$

The most significant determinant of PO_4^{3-} flux was concentration in the water, accounting for 55% of the variability observed. Dissolved oxygen was the next most significant determinant and accounted for 20% of the variability. Temperature accounted for none of the variability.

Discussion

This study concurs with reports that NH_4^+ release increases as a function of temperature but θ is lower than observed elsewhere (Bowden, 1984; Hale, 1975) and temperature explains a smaller fraction of the variability in flux than in other investigations (Boynton *et al.*, 1980; Hale, 1975). The disparate correlations of temperature and flux here and elsewhere may be due to differences in study objectives and designs. Measurements in Gunston Cove were conducted on sediments collected in a ten-week period and subjected to temperature manipulation. This program isolated the effect of temperature on the biological and chemical processes active in the sediments at the time of collection. Flux measures by Hale (1975) and by Boynton *et al.* (1980) were conducted in situ over seven to ten-month periods at ambient temperatures. Their flux-temperature correlations reflect not only temperature effects but also seasonal trends in sediment characteristics such as faunal density and input of organic detritus. In in-situ studies temperature serves as a single variable into which numerous influences are combined and high correlations of temperature and flux may be noted.

Substitution of appropriate parameters into Equation 1 indicates NH_4^+ release is enhanced by almost 70% as DO is reduced from 8 mg L^{-1} to zero (Fig. 1). Other studies have reported tripling of NH_4^+ release due to anoxia (Fillos & Molof, 1972) or else no effect (Fillos & Swanson, 1975). One mechanism that accounts for enhanced release at low DO concentrations is suppression of nitrification in the sediments. Ammonium that is no longer nitrified is released to the water. Enhancement of NH_4^+ release at low DO is also due to different metabolism of aerobic and anaerobic organisms. Experiments on flooded

soils indicate more NH_4^+ is released per unit of carbon decomposed by anaerobic bacteria than by aerobic bacteria (Tusneem & Patrick, 1971).

Forty-six percent of the variability of NH_4^+ flux remains unexplained. A residual of this magnitude suggests the existence of a significant influence not considered in the empirical model. Testing the hypothesis that NH_4^+ flux is influenced by NH_4^+ concentration in the water indicates no significant relationship. Sediment NH_4^+ release has been shown to be dependent upon the rate of input of organic matter (Kelly & Nixon, 1984), a factor not considered here. Variations in the availability of organic matter may account for the large coefficients of variation obtained in repeat incubations. If the unexplained variability in NH_4^+ release is related to organic matter input, a record or prediction of particulate organic matter in the water column may be useful in the prediction of sediment NH_4^+ release.

An increase in sediment uptake of NO_3^- as a function of water-column concentration has been noted in several systems (Boynton *et al.*, 1980; Boynton & Kemp, 1985; Cerco, 1985). In this study, uptake was linearly proportional to concentration. Other investigators (Messer & Brezonik, 1984; Nedwell, 1982; Van Kessel, 1977a) have described the effect of concentration on flux in terms of a rate-saturation equation. Half-saturation concentrations have been found in the range 3 to 50 mg L^{-1} . In this study, NO_3^- concentrations were generally 2 mg L^{-1} or less. Employing these concentrations in the reported rate-saturation equations yields functions that are approximately linear, indicating that the functional forms in this and other studies do not conflict.

Examination of the proportionality constant 'a' that relates NO_3^- flux to concentration in the overlying water, suggests uptake is affected by treatment of the sediments. The linear portion of Nedwell's (1982) data, obtained using intact sediment cores, indicates 'a' $\approx 500 \text{ mm day}^{-1}$. The data of Van Kessel (1977b), derived from observations of NO_3^- in a drainage ditch indicate 'a' = 130 mm day^{-1} . These values correspond in magnitude to 'a' = 110 mm day^{-1} obtained here

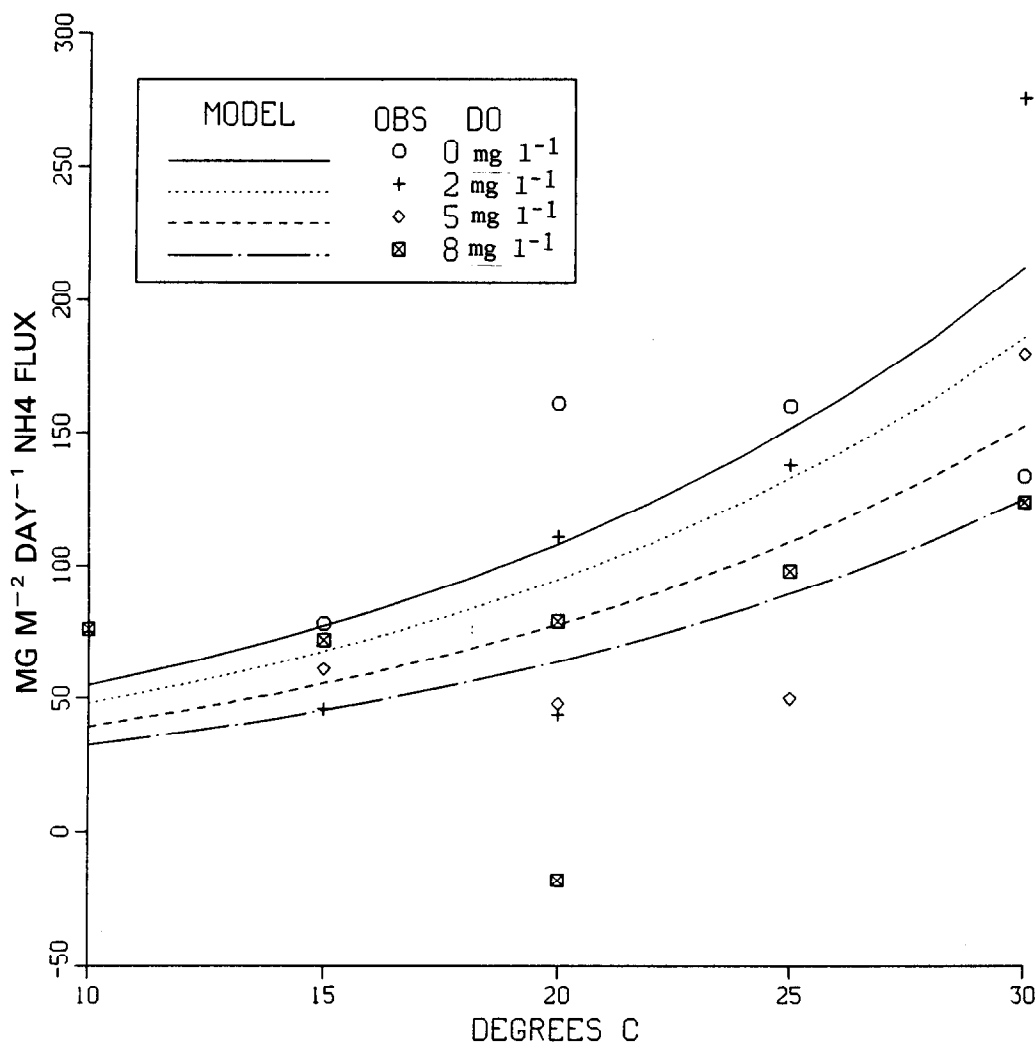


Fig. 1. Observed and modelled effects of temperature and DO on sediment NH_4^+ release.

using intact sediment cores. Van Kessel (1977a) also obtained NO_3^- flux versus concentration data employing sieved, homogenized sediments. The linear portion of these data indicate 'a' $\approx 5 \text{ mm day}^{-1}$. The proportionality constant obtained using undisturbed sediments is 20 to 100 times larger than the constant obtained using homogenized sediments.

The parameter θ derived here is within the range of values reported in other lab studies (Jenkins & Kemp, 1984; Klapwijk & Snodgrass, 1982; Messer & Brezonik, 1984; Seitzinger *et al.*,

1984). It is noteworthy that two in-situ studies (Cercio, 1985; Hale, 1975) found no effect of temperature on NO_3^- flux. As with NH_4^+ , disparate flux-temperature correlations in vitro and in situ may be due to in-situ variations in sediment properties. Contrary to NH_4^+ , however, NO_3^- flux shows a higher correlation with temperature in the laboratory than in situ.

Substitution of appropriate parameters into Equation 2 indicates sediment NO_3^- uptake almost doubles under anoxic conditions compared to uptake under a water column saturated

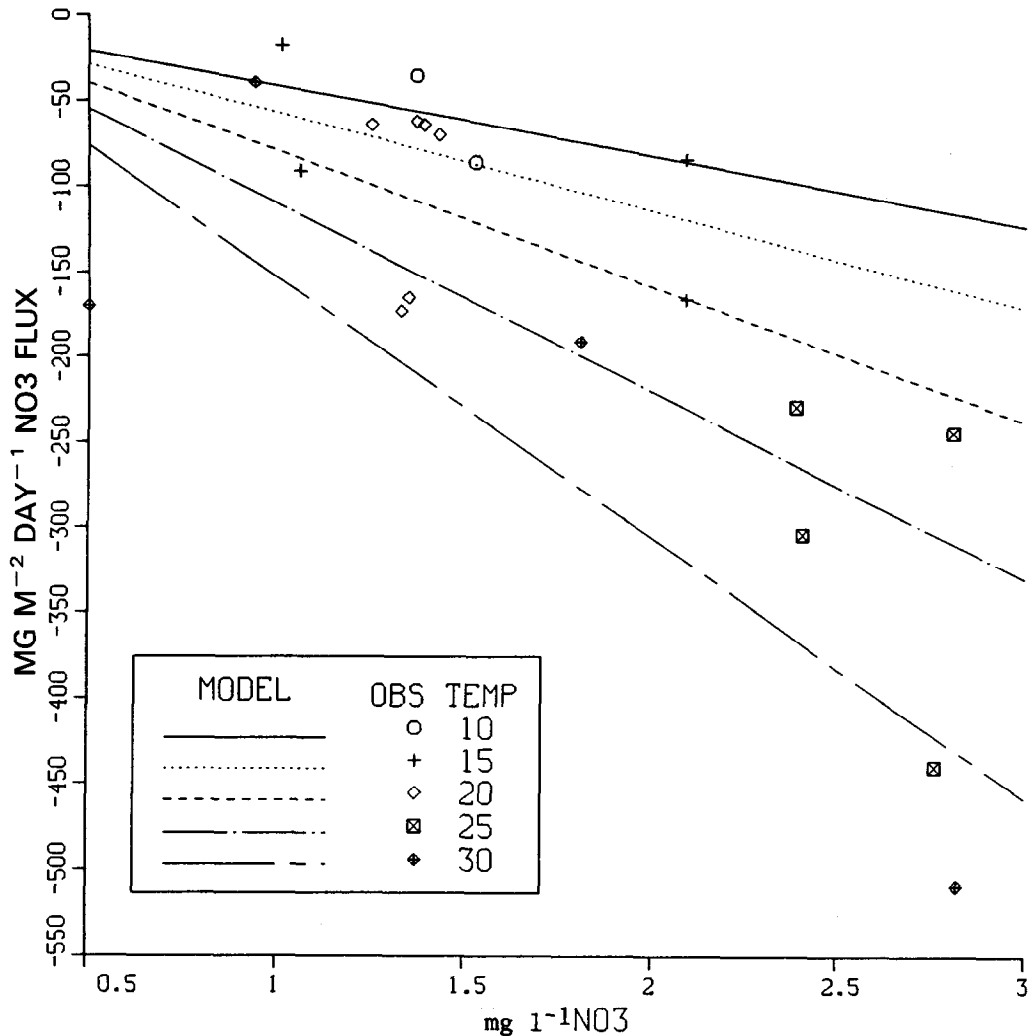


Fig. 2. Observed and modelled effects of concentration and temperature on sediment NO_3^- uptake. Observations collected at 0 to 8 mg L^{-1} DO. Modelled uptake computed at 4 mg L^{-1} DO.

with DO (Fig. 2). The enhanced uptake may be due to increased denitrification as DO becomes unavailable. The ratio of anoxic to oxic uptake noted here is at the upper range of reported values (Jacobsen & Jorgensen, 1975; Van Kessel, 1977a). Several system characteristics may affect the enhancement of NO_3^- uptake under anoxic conditions. Systems in which the rate of NO_3^- reduction is initially low relative to the rate of oxygen reduction, in which sufficient NO_3^- is available in the water column, and in which the diffusion of NO_3^- through the sediments is rapid,

will exhibit high ratios of enhancement under anoxic conditions. Systems in which the reduction of NO_3^- is initially large compared to the reduction of oxygen or in which the supply of NO_3^- to the sediments is limited, will exhibit low enhancement of sediment NO_3^- uptake under anoxic conditions.

The primary variable affecting PO_4^{3-} flux in these experiments was concentration in the water column. Under oxic conditions, PO_4^{3-} moved into or out of the sediments depending on PO_4^{3-} concentration. The experiments indicate the exist-

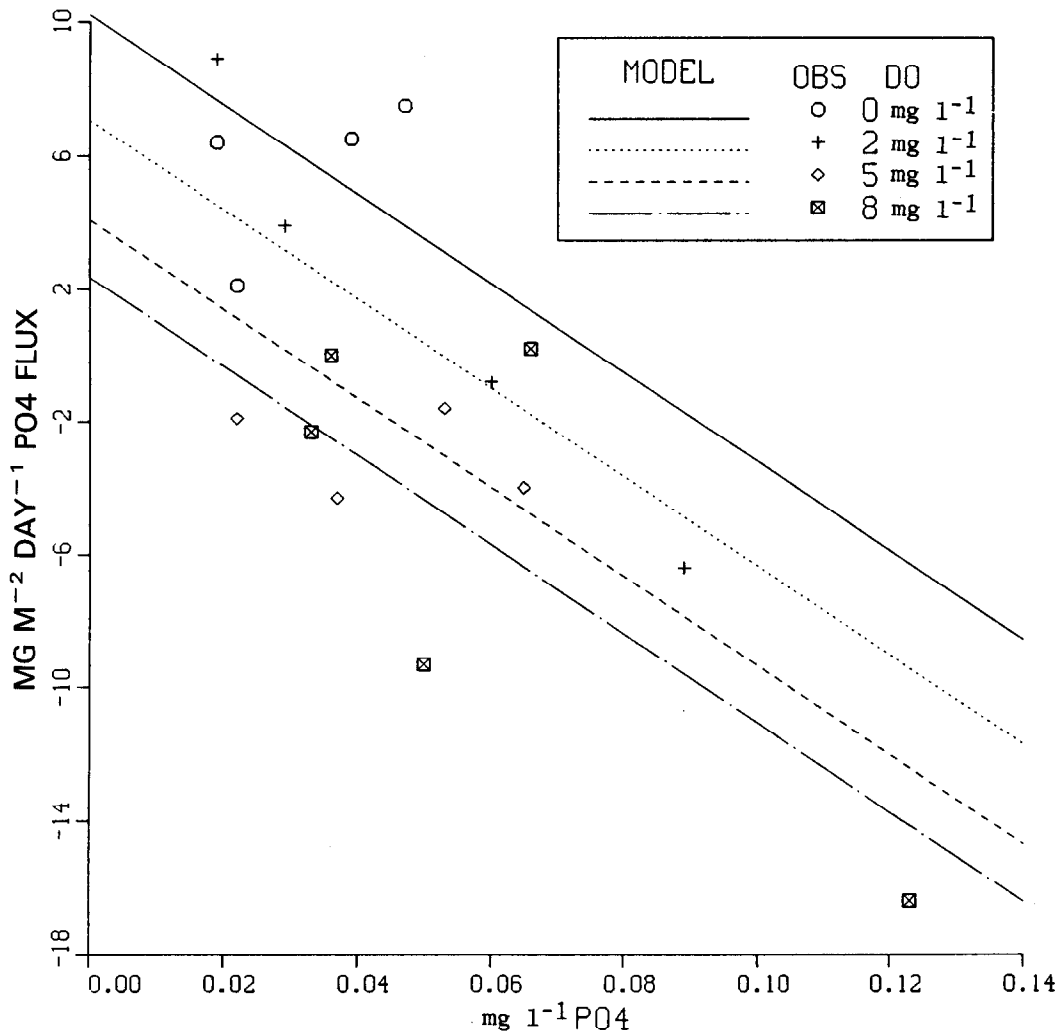


Fig. 3. Observed and modelled effects of concentration and DO on sediment PO_4^{3-} flux.

ence of a concentration at which PO_4^{3-} in the sediments is in equilibrium with the water column. When PO_4^{3-} in the water exceeds the equilibrium concentration, PO_4^{3-} moves into the sediments. When PO_4^{3-} in the water column is below the equilibrium concentration, PO_4^{3-} moves out of the sediments (Fig. 3). Solution of Equation 3 indicates that at 8 mg L^{-1} DO, the equilibrium concentration is $0.018 \text{ mg L}^{-1} \text{ PO}_4^{3-}$. The existence of an equilibrium PO_4^{3-} concentration in Gunston Cove is consistent with findings in other freshwater (Twinch & Peters, 1984) and brackish systems (Pomeroy *et al.*, 1965).

The model proposed here indicates that the equilibrium concentration is dependent on DO. As DO decreases, PO_4^{3-} is released to the water column until a new equilibrium is established. This principle is consistent with experiments conducted on lake sediments (Fillos & Biswas, 1976). Under oxic conditions, the sediments were a sink of phosphorus. Under anoxic conditions, the sediments were a source of phosphorus until the water column attained 1 to $1.4 \text{ mg L}^{-1} \text{ PO}_4^{3-}$. In that concentration range, no more phosphorus was released. The equilibrium concentration under anoxic conditions derived here, 0.07 mg L^{-1}

PO_4^{3-} , is much less than the concentration noted in that series of experiments, however.

Equation 3 indicates that anoxia in the water column increases sediment PO_4^{3-} release by a factor of four compared to flux under a DO-saturated water column containing no PO_4^{3-} . Other studies indicate an increase of phosphorus release of from two times to infinity under anoxic conditions compared to oxic conditions (Bates & Neafus, 1980; Fillos & Molof, 1972; Fillos & Swanson, 1975; Holdren & Armstrong, 1980; Mawson *et al.*, 1983; Nixon *et al.*, 1980). The extreme ratios of anoxic to oxic conditions occur in systems that release little or no phosphorus into an oxic water column. In systems that generally release phosphorus, the anoxic to oxic ratio is roughly five or ten to one. Gunston Cove sediments are at the lower end of this range.

Absence of influence of temperature on PO_4^{3-} flux, noted here, is supported by a series of observations on lake sediments (Andersen, 1974) but conflicts with the results of numerous other studies. Sediments have been noted to take up phosphorus at low temperatures and release it at high temperatures (Fisher *et al.*, 1982) or else to release phosphorus at a rate that is enhanced as temperature increases (Boynton *et al.*, 1980; Fillos & Swanson, 1975; Hale, 1975; Nixon *et al.*, 1980). The in-situ studies may overemphasize the influence of temperature on phosphorus flux due to the covariance of temperature with other factors but laboratory studies also demonstrate a temperature effect. The influence of DO and lack of influence of temperature suggest that interactions of iron and PO_4^{3-} dominate sediment-water PO_4^{3-} exchange in Gunston Cove. Holdren and Armstrong (1980) found that PO_4^{3-} exchange with iron-rich sediments was dominated by redox conditions while PO_4^{3-} exchange with calcareous sediments was dominated by temperature effects.

The tendency for Gunston Cove sediments to take up PO_4^{3-} under ambient conditions ($\text{DO} > 5 \text{ mg L}^{-1}$) conflicts with the notion that sediment phosphorus release supports algal blooms in the Cove (Thomann *et al.*, 1985). Sediment PO_4^{3-} release in these experiments may

have been limited by the pH prevalent in 1984, 7.2 to 8.6. The pH during algal blooms may exceed 9.5 and pH-induced PO_4^{3-} release has been proposed to occur during these blooms (Thomann *et al.*, 1985). Lijklema (1977) indicated the sorption capacity of ferric hydroxide declines as pH increases. If, as suggested, PO_4^{3-} flux in Gunston Cove is dominated by interactions of Fe^{3+} and PO_4^{3-} , then sediment PO_4^{3-} release should increase as a function of pH due to diminished sorption of PO_4^{3-} on iron complexes.

Conclusions

Interactions of sediment nutrient fluxes and conditions in the overlying water cannot be described exclusively by reference to published reports. If site-specific observations are available, NO_3^- and PO_4^{3-} flux may be well-described by empirical relationships fit to the data. Flux of NH_4^+ is least susceptible to description by empirical means. Incorporating a record of organic input is suggested as an improvement to the model proposed here. Disparities in θ evaluated in the laboratory and in situ indicate that θ is best evaluated by incubating sediments collected seasonally at in-situ temperatures.

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