Reports

Transformations of Nitrogen in a Polluted Estuary: Nonlinearities in the Demand for Oxygen at Low Flow

Abstract. *Oxidation of sewage ammonium in the Potomac River is described in terms of a simple kinetic model, with growth of nitrifying bacteria limited by the supply of ammonium ion. The oxidation rate varies inversely with freshwater inflow, and the associated demand for oxygen varies as the inverse square of the freshwater inflow rate. Similar behavior is observed for the Delaware River. The model accounts for the observed concentrations of ammonium and nitrous oxide.*

Many large cities are located on rivers or estuaries. Much of the associated sewage, treated and untreated, is released into these systems, often with deleterious effects on local aquatic life. The impact on water quality depends on a series of complex chemical transformations that in general are not well defined. This report focuses on a nonlinear aspect of the chemistry affecting nitrogen. The rate at which ammonium is oxidized in estuaries is shown to vary inversely with freshwater inflow. Oxidation of NH_4^+ represents an important demand for dissolved oxygen. The concentration of O_2 may respond in an unexpected fashion to changes in stream flow and variations in the composition of effluent. The complexity of nitrogen chemistry has implications for water management and must be recognized in future engineering models for polluted estuaries.

Our analysis is based on 10 years of data for the Potomac River estuary (*1*). The conclusions are more general, however, since they are also valid for the Delaware River estuary (*2*). The Potomac is particularly simple in that the sewage supply of nitrogen may be regarded as a constant point source of NH_4^+ (~ 200 g of nitrogen per second) (*1*, *3*). In contrast, the river's rate of flow varies widely, even during summer. Between July and September, flow ranges from ~ 20 to > 250 m^3 sec^{-1}, with a mean of ~ 130 m^3 sec^{-1} (*4*).

Ammonium is removed by nitrifying bacteria, which mediate the reaction

$$NH_4^+ + \frac{3}{2}O_2 \rightarrow NO_2^- + 2H^+ + H_2O \quad (1)$$

where the free energy change $\Delta G = -59.3$ kcal $mole^{-1}$. Ammonium is also used by growing phytoplankton (*5*). Nitrifying bacteria are noted for their relatively slow growth and for inefficient use of the energy derived from reaction 1 (*6*, *7*). Nitrous oxide (N_2O) is a by-product of reaction 1, and, as discussed elsewhere (*8*, *9*), observations of N_2O may be used to obtain direct information on the rate of nitrification.

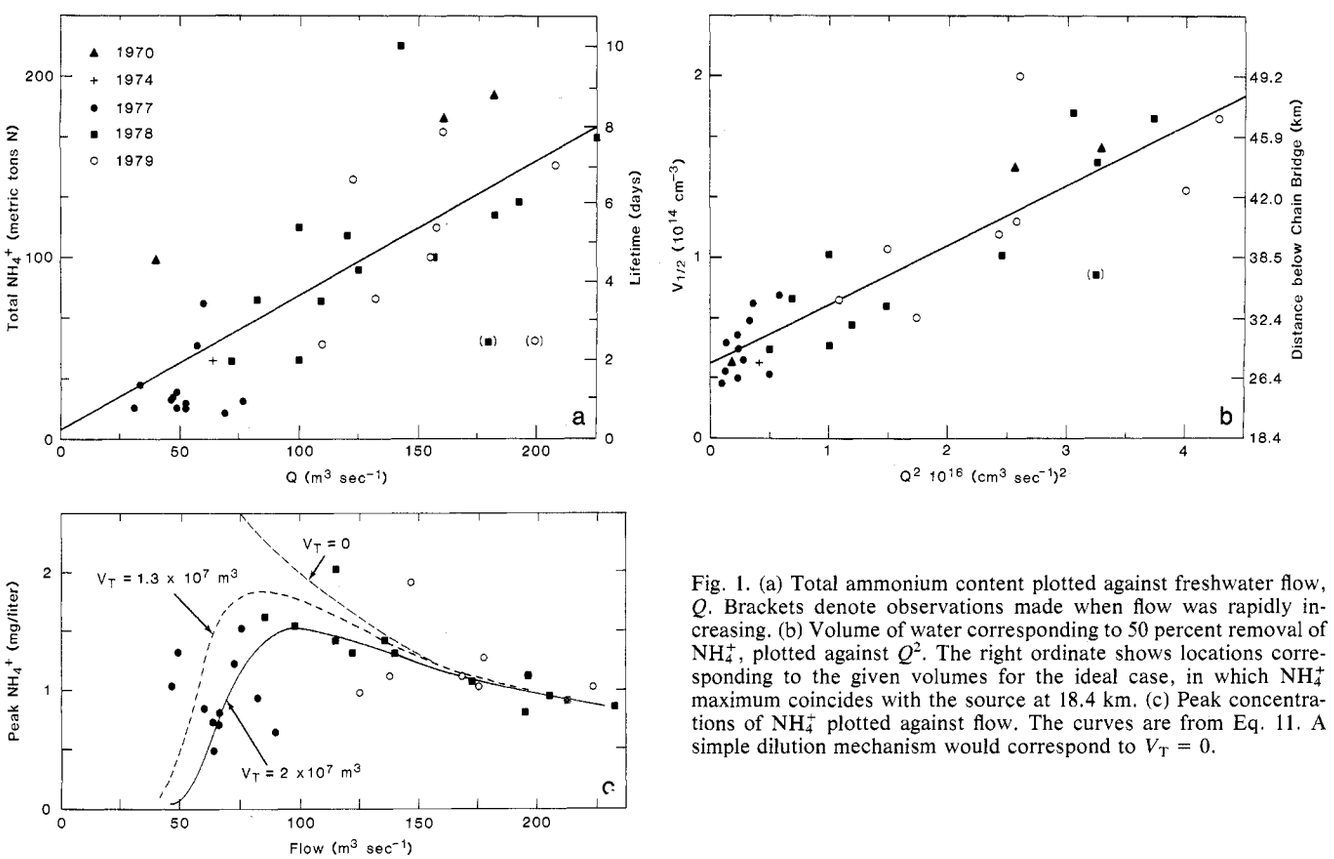

Fig. 1. (a) Total ammonium content plotted against freshwater flow, Q. Brackets denote observations made when flow was rapidly increasing. (b) Volume of water corresponding to 50 percent removal of NH_4^+, plotted against Q^2. The right ordinate shows locations corresponding to the given volumes for the ideal case, in which NH_4^+ maximum coincides with the source at 18.4 km. (c) Peak concentrations of NH_4^+ plotted against flow. The curves are from Eq. 11. A simple dilution mechanism would correspond to $V_T = 0$.

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Figure 1a illustrates the relation between river flow Q ($cm^3\ sec^{-1}$; 1 $m^3\ sec^{-1} = 10^6\ cm^3\ sec^{-1}$) and total content of NH_4^+ observed in the Potomac for water temperatures between 23° and 31°C (*10*). If removal of NH_4^+ were described by a first-order rate constant ν (sec^{-1}), the NH_4^+ content N would be given by

$$N = q_N \nu^{-1} \tag{2}$$

where q_N is the input rate (*11*). The content of NH_4^+ appears to increase with stream flow Q (Fig. 1a). Thus the data are not consistent with Eq. 2 if we require that ν be constant. They may be accommodated, however, with a model in which ν is inversely proportional to Q.

The time rate of change of NH_4^+ due to reaction 1 may be described by

$$\frac{d[NH_4^+]}{dt} = -a\Gamma b(t)[NH_4^+] \tag{3}$$

where $[NH_4^+]$ denotes the concentration of NH_4^+ (grams of nitrogen per cubic centimeter; 1 mg liter $^{-1} = 10^{-6}$ g cm^{-3}), b is the concentration of bacteria (cm^{-3}), a is the average mass of NH_4^+ assimilated or oxidized by an individual cell from birth to division (2×10^{-11} g of nitrogen) (*7*), and Γ is the growth rate of a cell at unit concentration of NH_4^+ (sec^{-1} g^{-1} cm^3). Equation 3 assumes that the growth of bacteria is limited primarily by supply of NH_4^+ (*12*). The time rate of change of b is given by

$$\frac{db}{dt} = \Gamma[NH_4^+]b(t) \tag{4}$$

Denote the cumulative volume of water (cm^3) at point z downstream from the point of NH_4^+ input z_0 by $V(z)$. To the extent that we may ignore turbulent dispersion—justified except for the lowest flow rates (*13*)—then

$$\frac{dV}{dt} = Q \tag{5}$$

Thus time may be eliminated from Eqs. 3 and 4, permitting solution for $[NH_4^+]$ and $b(t)$ in terms of Q. The concentration of NH_4^+ is given by

$$[NH_4^+] = \frac{q_N}{Q}\,\frac{1+\alpha}{1+\alpha \exp\left[q_N\Gamma(1+\alpha)V(z)/Q^2\right]} \tag{6}$$

and

$$b(t) = b_0 + a^{-1}\{[NH_4^+]_0 - [NH_4^+]\} \tag{7}$$

where $\alpha = aq_b/q_N$. Here q_b is the input rate (sec^{-1}) of nitrifying organisms, b_0 ($= q_b/Q$) is the concentration of organisms at z_0, and $[NH_4^+]_0$ ($= q_N/Q$) is the concentration of NH_4^+ at z_0. Equation 6 implies that the peak concentration of NH_4^+ should occur at the source and should decline by half at a position corresponding to

$$V = V_{1/2} = \frac{Q^2}{q_N(1+\alpha)\Gamma} \ln(2 + \alpha^{-1}) \tag{8}$$

The total content of NH_4^+ is given by $N = Q\Gamma^{-1} \ln(1 + \alpha^{-1})$. This expression is consistent with the general behavior of the data presented in Fig. 1a. Oxygen is consumed at a rate proportional to $d[NH_4^+]/dt$, given by

$$\frac{d[NH_4^+]}{dt} = -\left(\frac{q_N}{Q}\right)^2 \Gamma(1+\alpha)^2 \frac{E}{(1+E)^2} \tag{3'}$$

with $E = \alpha \exp[q_N\Gamma(1+\alpha)V]$. Consumption of O_2 reaches a maximum immediately upstream of $V = V_{1/2}$, where demand for O_2 varies as $q_N{}^2Q^{-2}$—as can be seen by setting $E = 1$.

The expressions for $[NH_4^+]$, $b(t)$, and $d[NH_4^+]/dt$ contain two adjustable parameters, α (or aq_b) and Γ. The other variables [q_N, Q, and $V(z)$] can be obtained from public records (*3, 4*). Measurements of $V_{1/2}$ are summarized in Fig. 1b. The solid line represents a fit of the data to an expression of the form

$$V_{1/2} = AQ^2 + B \tag{8'}$$

where B is included to allow for effects of dispersion at low values of Q, and A, according to Eq. 8, is given by $[q_N(1+\alpha)\Gamma]^{-1} \ln(2 + \alpha^{-1})$. Data for total NH_4^+ content N were fitted to an expression of the form

$$N = A'Q + B' \tag{9}$$

where $A' = \Gamma^{-1} \ln(1 + \alpha^{-1}) = 0.75 \pm 0.14$, with $B' \cong 0$. The solid lines in Fig. 1, a and b, correspond to the values $\Gamma = 3.42$ and $\alpha = 0.1$ ($q_b \approx 10^{12}$ sec^{-1}). Values of N and $V_{1/2}$ depend only weakly on α: the value for α selected here was determined by considering individual profiles for NH_4^+ measured in August 1978 (Fig. 2a) (*14*). The value for Γ implies a doubling time for nitrifying bacteria of 2.3 days at an ammonium concentration of 1 mg of nitrogen per liter—consistent with growth rates observed for pure cultures in the laboratory (*6, 8*). The concentration of nitrifying organisms at z_0 is predicted to be about 10^4 cm^{-3} for $Q = 100$ m^3 sec^{-1}, rising to 3×10^4 cm^{-3} at $V_{1/2}$ and ultimately to 6×10^4 cm^{-3} (*15*).

We have assumed so far that NH_4^+ is removed mainly by reaction 1. The analysis could be generalized by suitable redefinition of $b(t)$ to account for uptake by phytoplankton. There are reasons to believe, on the other hand, that reaction 1 is indeed dominant, at least for the Potomac and Delaware rivers. Figure 2b shows profiles for N_2O obtained with a model in which the gas is assumed to form as a by-product of reaction 1 at a rate proportional to ammonia removal (Eq. 3'). The concentration n (μg $liter^{-1}$) of dissolved N_2O satisfies an equation of the form

$$\frac{d(n - n_e)}{dt} = fY \frac{d}{dt}[NH_4^+] - \frac{n - n_e}{\tau} \tag{10}$$

where n_e is the concentration in equilibrium with the atmosphere (about 0.35 μg $liter^{-1}$), f denotes the fraction of NH_4^+ removed by reaction 1, Y is the yield of N_2O associated with nitrification (moles of nitrogen as N_2O per mole of nitrite), and τ is a characteristic time for exchange of gas between water and air, about 6 days for the Potomac (*16, 17*). Figure 2b shows that the model gives excellent agreement with observation if fY is set equal to 2.5×10^{-3}. Yields Y measured in the laboratory range from 2×10^{-3} to 3.5×10^{-3} (*8*), and in situ determinations for the Potomac (*18*) average 3.3×10^{-3}. It appears that between 70 and 100 percent of NH_4^+ in the Potomac is removed by reaction 1. Observations of NO_2^- and NO_3^- are consistent with this view (*19*).

The model gives a satisfactory account for the distribution of NH_4^+ (Fig. 2a), except when stream flow is less than ~ 80 m^3 sec^{-1}. The discrepancy at low Q is to be expected, since the lifetime of NH_4^+ is very short in this case, often less than 2 days (Fig. 1a). The treatment of transport must be adjusted at short lifetimes to allow for rapid oxidation in the shallow embayment receiving the bulk of the sewage discharge. The assumption of a point source also must be modified as the width of the NH_4^+ distribution becomes less than 10 km. Our complete hydraulic and kinetic model (*13*) adequately accounts for these details.

The maximum concentration of NH_4^+ exhibits peculiar behavior at low flow (Fig. 1c). Peak concentrations increase as flow declines from 200 to 100 m^3 sec^{-1} but decrease below 100 m^3 sec^{-1}. Sewage ammonia is distributed by tidal currents over a volume, V_T, which is slightly larger than the volume of water flowing back and forth in a tidal cycle (1.3×10^7 m^3 at the Blue Plains Regional

Sewage Plant). The mean residence time for NH_4^+ in V_T is V_T/Q, and the concentration is given by

$$[NH_4^+]_0 = \frac{(q_N/Q)(1+\alpha)}{1+\alpha \exp[q_N\Gamma(1+\alpha)V_T/Q^2]} \quad (11)$$

This expression reproduces the observed behavior of NH_4^+ shown in Fig. 1c, with Γ and α as derived above and with $V_T \simeq 2 \times 10^{13}$ m^3. The reduction in NH_4^+ at low flow reflects intense activity of nitrifying bacteria near the plant. The oxidation rate may be predicted by using Eq. 3′.

The effect of temperature on nitrification may be distinguished from the influence of flow by means of observations made in fall and spring (water temperature, 17° to 21°C). Analysis of these data suggests that the rate of nitrificaton is lowered by a factor of ~ 1.6 ± 0.4 for a reduction in water temperature of 10°C. Reported values for this temperature coefficient range between 1.5 and 2.2 (*6*) for cultures of nitrifying bacteria.

In summary, the biogeochemistry of nitrogen in the Potomac River can be described in terms of a simple kinetic model incorporating representative data for N_2O and NH_4^+. The rate of nitrification varies inversely with flow rate. The rate for O_2 consumption due to nitrification reaches a maximum proportional to $(q_N/Q)^2$, where q_N is the rate for input of NH_4^+. The influence of flow rate on the oxidation of ammonium is unexpected, and must be recognized in assessing projects that affect river flow and nitrogen input.

S. C. WOFSY
M. B. MCELROY
Center for Earth and Planetary Physics, Harvard University, Cambridge, Massachusetts 02138
J. W. ELKINS
National Bureau of Standards, Gas and Particulate Science Division, Washington, D.C. 20234

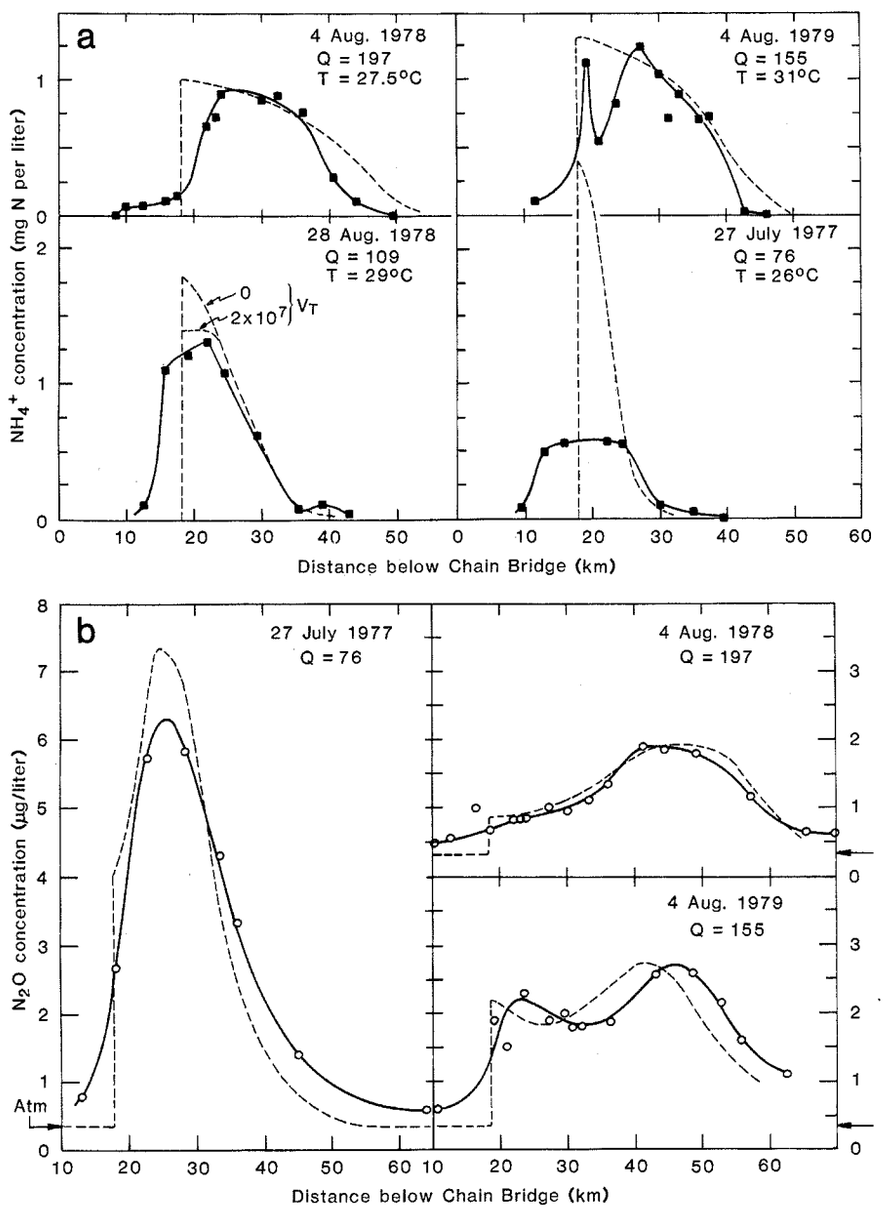

Fig. 2. Distributions of NH_4^+ (a) and N_2O (b) in the Potomac for a range of flow rates. Model results (dashed lines) were calculated for $\Gamma = 3.4$ sec^{-1} g^{-1} cm^3, $\alpha = 0.1$, $fY = 2.5 \times 10^{-3}$, and $\tau = 6$ days, except for 4 August 1979, where $\alpha = 0.05$. Arrows denote the concentration of N_2O in equilibrium with the atmosphere. The source of N_2O from chlorination is 0.06 g sec^{-1}, except for 4 August 1979, when it was 0.2 g sec^{-1} (*18*). Production of N_2O from nitrification is 0.5 g sec^{-1} ($= f\,Y\,q_N$). The influence of a finite initial mixing volume (see Fig. 1c) is shown for 28 August 1978.

References and Notes

1. L. J. Clark and N. A. Jaworski, *Environ. Prot. Agency (Annapolis Field Office) Tech. Rep. 37* (1972); L. J. Clark and S. E. Roesch, *Environ. Prot. Agency (U.S.) Rep. 903/9-78-008* (1978); J. Prendergast, private communication. Our own data (1977–1979) were acquired as discussed in M. B. McElroy, J. W. Elkins, S. C. Wofsy, C. E. Kolb, A. P. Duran, and W. A. Kaplan [*Limnol. Oceanogr.* **23**, 1168 (1978)]. Data include concentrations of NH_4^+, NO_2^-, NO_3^-, total N, N_2O, temperature, and salinity. The estuary is vertically homogeneous in the region of interest. Most samples were from the main channel at a depth of 0.5 m.
2. We have 3 years of data (1978–1980) for the Delaware River (in preparation).
3. E. R. Jones, Wastewater Division, Government of the District of Columbia, monthly reports (1977–1979). The Blue Plains Regional Sewage Plant, located 18 km below the head of tide (Chain Bridge), acounted for 80 percent of the sewage nitrogen input to the Potomac. Most of the remaining nitrogen was released at Arlington (15 km below Chain Bridge) and Alexandria (22 km). Tidal oscillations extend 8 km per half-cycle, and individual sources are usually indistinct. About 80 percent of the nitrogen is released as NH_4^+ and 20 percent as organic nitrogen, which is rapidly converted in the river to NH_4^+ (*1*). Blue Plains commenced advanced treatment in 1980. Most of the nitrogen is now converted to NO_3^- or NO_2^- before being released into the river.
4. U.S. Geological Survey, Towson, Md. (courtesy of M. Liss). Flow of sewage is 15 m^3 sec^{-1}.
5. R. W. Eppley, J. N. Rogers, J. J. McCarthy, *Limnol. Oceanogr.* **14**, 912 (1969); W. A. Harvey and J. Caperon, *Pac. Sci.* **30**, 329 (1976).
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8. T. J. Goreau, W. A. Kaplan, S. C. Wofsy, M. B. McElroy, F. W. Valois, S. W. Watson, *Appl. Environ. Microbiol.* **40**, 526 (1980).
9. J. W. Elkins, S. C. Wofsy, M. B. McElroy, W. A. Kaplan, in *Nutrient Enrichment in Estuaries*, B. Nielsen, Ed. (Humana, Clifton, N.J., in press).
10. Total content of NH_4^+ is defined as $N = \int_{z_1}^{z_2} [NH_4^+]\,dV$, where z_1 is upstream of the major inputs and z_2 is the point where $[NH_4^+]$ is reduced to 0.1 mg liter^{-1}. Excess NH_4^+ occasionally appears far downstream due to collapse of phytoplankton blooms. This secondary peak of NH_4^+ was excluded from the calculation.
11. The source of NH_4^+ was quite steady when averaged over several days (*3*) and did not respond to local precipitation events, as evidenced by low concentrations of NH_4^+ above the sewage plants. Ablation of estuarine sediments is controlled by tidal currents, since mean flow velocity is less than 10 percent of tidal currents for $Q < 200$ m^3 sec
12. The complete growtn equations for nitrifying bacteria may be written as $db/dt = -1/a$ $d[NH_4^+]/dt - Mb$ and $d[NH_4^+]/dt = -a\Gamma b[NH_4^+]$ $(1 + [NH_4^+]/X_s)^{-1}$ [F. E. Stratton and P. L. McCarty, *Environ. Sci. Technol.* **1**, 405 (1967)], where Mb accounts for death and predation and X_s is the half-saturation constant. We assume Mb is negligible during rapid growth. Laboratory studies (*6*, *7*) suggest that X_s is greater than

the highest concentration of NH_4^+ typically found in the Potomac (~ 1.5 mg liter^{-1}). Growth is limited therefore by substrate [NH_4^+], and the equations reduce to Eqs. 3 and 4. The behavior of NH_4^+ in the Potomac and Delaware rivers implies that microbial populations move with river flow, whereas in shallow streams nitrifying activity occurs at the bottom [T. J. Tuffey, J. V. Hunter, V. A. Matulewich, *Water Res. Bull.* **10**, 555 (1974)].

13. The validity of Eq. 5 was verified by detailed analysis with a complete hydraulic and kinetic model.
14. The value $\alpha = 0.1$ gave the best fit for 1977 and 1978; 0.05 is optimal for 1979.
15. The number of active nitrifying bacteria should be measurable in principle, but existing methods (most probable number) appear unreliable. Numbers of nitrifying bacteria predicted here are comparable to values determined by E. J. C. Curtis, K. Durrant, and M. M. I. Karman [*Water Res.* **9**, 255 (1975)] for polluted estuaries in England.
16. D. E. Hammond and C. Fuller [*Eos* **61**, 1003 (1980)] estimated τ in the range 4.5 to 7 days by using radon deficit measurements. The engineering formula $\tau = (d^3/(Dv))^{1/2}$ gives a similar value, $\tau = 5.7$ days, with depth $d = 450$ cm, tidal velocity $v = 15$ cm sec^{-1}, and diffusion coefficient $D = 2.5 \times 10^{-5}$ cm^2 sec^{-1} (*1*).
17. Small quantities of N_2O are formed during chlorination of sewage—typically ~ 10 percent of the source from nitrification (0.06 g sec^{-1} compared to 0.5 g sec^{-1} from nitrification). Slightly larger quantities of N_2O (0.2 g sec^{-1}) were produced as a result of heavy chlorination in 1979 [R. J. Cicerone, J. D. Shetter, S. C. Liu, *Geophys. Res. Lett.* **5**, 173 (1978); W. A. Kaplan, J. W. Elkins, C. E. Kolb, M. B. McElroy, S. C. Wofsy, A. P. Duran, *Pure Appl. Geophys.* **116**, 423 (1978)].
18. Yields of N_2O from nitrification in the Potomac were estimated also by observing the time courses of N_2O, O_2, NH_4^+, NO_2^-, and NO_3^- in water samples placed in sealed vessels. Observations at 0, 7, 24, and 48 hours showed parallel increases in N_2O and (NO_2^- + NO_3^-), with N_2O yields between 3.0×10^{-3} and 3.7×10^{-3} mole of nitrogen per mole of NO_2^- on 5 August 1979 and between 1.8×10^{-3} and 4×10^{-3} on 17 August 1980.
19. The concentration of (NO_2^- + NO_3^-) increases downstream of sewage plants as NH_4^+ is oxidized. On average, the peak concentration of (NO_2^- + NO_3^-) accounts for 54 ± 14 percent of the total nitrogen source. However, a much higher fraction of NH_4^+ may be removed by reaction 1, since oxidized species (NO_2^- and NO_3^-) are rapidly consumed in the estuary (*9*).
20. We are indebted to T. Flaherty for invaluable discussions and to C. Spivakovsky, W. Kaplan, L. Hashimoto, A. Duran, C. Kolb, T. Goreau, G. Soffen, J. Ledwell, P. Tolbert, A. Appiah, S. Troian, S. Frankel, and K. Buttleman for help in sampling and analysis. Supported by NSF grant DEB79-20292, EPA grant R807081010, and NASA grant NASW 2952 to Harvard University.

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Lapita Colonization of the Admiralty Islands?

Abstract. *Archeological research in the Admiralty Islands provides evidence of occupation by 3500 years ago and suggests settlement by obsidian-using maritime colonists, whose Lapita pottery style underwent gradual modification within the Admiralties.*

Excavations at Kohin Cave (*1*), Admiralty Islands, Manus Province, Papua New Guinea, have produced pottery and Lou Island obsidian throughout well-stratified deposits. Four sherds from lower stratigraphic layers are decorated with dentate-stamped impressions, distinctive of the Lapita style (*2, 3*). Lou Island, in the Admiralties, is one source of obsidian found in Lapita sites further east (*4, 5*). The dentate-stamped sherds from Kohin Cave are the first evidence of a cultural association between the Lapita complex and an assemblage of comparable age from the Admiralty Islands. The sherds are in a secure stratigraphic context, from layers bracketed by carbon-14 dates. The evidence from Kohin Cave suggests that these sherds and others from the lower layers represent an early phase in a local developmental sequence, beginning with the settlement of the Admiralties by seagoing colonists whose pottery was in the Lapita style.

Two excavations of Kohin Cave, on the southeast coast of Manus Island, in 1978 (*1*) and 1979 covered 11 m^2. Excavation followed depositional strata, and disturbances and discontinuous lenses were isolated. The stratigraphy of the two excavations correlates well. Most layers, although varying in thickness, are continuous throughout. Excavation techniques and layer correlations allow the provenance of cultured materials and their association with dated samples to be stated with confidence.

The deposits, in ten major layers, all contain cultural material and are built up on a bedrock base at an average depth of 1 m below the surface. Two layers have special significance. Layer 4, continuous throughout the excavated area, has a surface defined by several hearths and scattered ash and charcoal, indicating a period of occupation. Layer 10, a shell midden, is built up on bare rock. Carbon-14 dates have established the chronology (Table 1). The layer 4 samples are from opposite ends of the 1978 excavation and date cultural features in that layer. The charcoal of the layer 5 sample is not as clearly the product of human action as are other samples but is stratigraphically associated with cultural material. The layer 10 sample (from a small species of *Tridacna*) dates the surface of that layer; stratigraphic evidence indicates that layer 10 was deposited rapidly. There is no evidence to suggest a chronological hiatus between layer 10 and those above it.

Sherds and obsidian flakes and spalls occur throughout the layers, but there were no other significant artifacts. The obsidian derives from several sources on Lou Island (*6*). Neither pot forms nor fabric types can be satisfactorily distinguished from the sherds, which are mostly very small and eroded or chemically altered (Table 2). In layer 10, sherds are rare, even taking into account the much smaller volume excavated; however, that layer may have been deposited virtually during a single cultural event.

Distinctive sherds (rims and decorated pieces) provide a basic ceramic sequence, of which the salient features are as follows: (i) there is overall continuity of rim forms and decoration; (ii) several new rim forms and elaborations of decoration appear in layer 4; (iii) the four dentate-stamped sherds derive from layers 7, 8, and 9; and (iv) the layer 10 sherds are not distinctive. Figure 1 shows typical decorated sherds, including all the decorated body sherds from layers 7 through 9. Because of fragmentation and chemical alteration, the four dentate-stamped sherds cannot be clearly associated with others from the same layers on the basis of either formal or fabric identity. Each sherd seems to bear a different motif, each executed with a tool of different dimensions. The other four decorated body sherds and decorated rims from layers 7 through 9 have shell impressions or linear incisions. Such decoration is not inconsistent with the Lapita style (*2, 3, 7*), but it is not itself distinctive and occurs throughout the Kohin sequence. From stratigraphic associations and position in the ceramic

Table 1. Carbon-14 (half-life 5570 ± 30 years) dates from Kohin Cave. The age of sample ANU-2248 is corrected to the equivalent charcoal age (*15*).

Number	Sample	Layer	Age (years)
ANU-2089	Charcoal	4	2070 ± 120
ANU-2215	Charcoal	4	1910 ± 90
ANU-2212	Charcoal	5	2310 ± 120
ANU-2248	Shell	10	3450 ± 100

Table 2. Numbers and types of sherds in various stratigraphic layers of Kohin Cave. Totals include all sherds, and rims include both plain and decorated sherds; decorated refers to body sherds only. Approximate densities were derived from the area excavated and the estimated average thickness of the layer.

Layer	Sherd count			Density (m^3)
	Rims	Decorated	Total	
1 to 3	30	17	826	376
4	27	23	591	358
5 and 6	9	1	259	173
7 to 9	28	8	883	370
10	0	0	8	24

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