with a shifted time origin that accounts for the initial condition J^*, just as predicted by Eq. 10. Extrapolating this model, it is expected that the rate of current decay present in the sample after the quench at 80 s shown in Fig. 4 is 1.0 ppm/s. This level of stability would be obtained without a quench only after 20,000 s at 77.5 K, according to Eq. 7.

Therefore, we conclude that the relaxation of magnetic shielding current can be greatly reduced and effectively eliminated if the superconductor is operated at current levels slightly below the critical current density. Since the reduction of the relaxation depends on the operating current reduction level η with a very large power exponent n, such improvements can be made without great sacrifice of the utility of the critical current density. Thus, the relaxation of the magnetic shielding current in the YBCO material is not a serious problem for persistent current applications. A convenient way of achieving this condition, as we have demonstrated, is to simply reduce the operating temperature slightly after the sample enters its critical state.

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Sedimentary 24-*n*-Propylcholestanes, Molecular Fossils Diagnostic of Marine Algae

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Certain C_{30}-steranes have been used for identifying sedimentary rocks and crude oils derived from organic matter deposited in marine environments. Analysis of a C_{30}-sterane from Prudhoe Bay oil indicates that these C_{30}-steranes are 24-*n*-propylcholestanes that apparently are derived from precursor sterols 24-*n*-propylidene-cholesterols and 24-*n*-propylcholesterol. These widely occurring sterols are biochemically synthesized in modern oceans by members of an order (Sarcinochrysidales) of chrysophyte algae. These data thus imply that C_{30}-sterane biomarkers in sedimentary rocks and crude oils have a marine origin. Screening of a few organic-rich sedimentary rocks and oils from throughout the Phanerozoic suggests that these C_{30}-steranes first appeared and, therefore, their source algae evolved between Early Ordovician and Devonian.

A SERIES OF C_{30}-STERANE EPIMERS containing 11 C atoms in the side chain has been empirically established as a widespread fossil marker for marine organic input to sedimentary rock and petroleum (*1–3*). This concept has been challenged recently (*4*), and some possible C_{30}-sterane precursors have been tentatively identified in lacustrine sediments (*5, 6*). In order to evaluate the origins of these steranes and identify them, we have studied a C_{30}-sterane sourced from marine shale present in a Prudhoe Bay petroleum (*2*).

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This C_{30}-sterane (12 ppm in the Prudhoe Bay oil) was isolated by a lengthy procedure. The oil was fractionally distilled under vacuum on a spinning band column to provide a narrow-boiling fraction containing the C_{30}-steranes. The fraction was separated by preparative silica gel chromatography, and the saturate fraction was deparaffinated with 5A molecular sieves. Key steps were the removal of certain terpanes (that is, hopanes) with 13X molecular sieves (*7*) and the removal of sterane epimers by chromatography on alumina (*8*). These two steps removed the bulk of the polycyclic biomarkers with physical properties similar to those of the target sterane. Repeated, nonaqueous reversed-phase, high-performance liquid chromatography (HPLC) provided 0.6 mg of 65% pure material. This sample was suitable for comparisons by nuclear magnetic resonance (NMR) with the synthetic materials because all contaminating components were minor constituents, each composing less than 5% of this mixture.

As the standards, we prepared the 5β and 5α epimers of 24-*n*-propylcholestane (structures **1bA** and **1aA** in Fig. 1) and 24-isopropylcholestane (structures **1bD** and **1aD**) by partial synthesis (Fig. 2) as described (*9–12*). Comparisons of the synthetic standards with the isolated C_{30}-sterane by gas chromatography–mass spectrometry (GCMS), ^{13}C NMR, and H NMR indicated that it is (24*R* + 24*S*)-24-*n*-propylcholestane (**1aA**). Its mass spectrum (Fig. 3) is identical to that of synthetic **1aA** and shows the expected D-ring cleavage having a mass-to-charge ratio (m/z) 217 and B-ring cleavage (m/z 304) ions characteristic of a sterane with a $C_{11}H_{23}$ side chain. The ^{13}C NMR spectrum of the isolated material was of sufficient strength to show that it is identical with **1aA**. Both contained a peak at 14.6 ppm diagnostic for the presence of the *n*-propyl group [predicted 14.4 ppm on the basis of Lindeman-Adams parameters (*13*)]. Finally, a fingerprint comparison of 300-MHz H NMR spectra in the methyl resonance region confirmed the structure assignment (Fig. 4).

Steranes are typically found in thermally mature sediments and petroleums as mixtures of stereoisomers at the C-5, 14, 17, 20, and 24 positions (*12, 14*). In order to confirm that the C_{30}-steranes (Fig. 5A) belong to the 24-*n*-propylcholestane series, we prepared an isomerizate by treatment of 24-*n*-propyl-5β-cholestane (**1bA**) with Pd/C at 260°C for 68 hours (*15*). This procedure produced a petroleum-like distribution of sterane epimers (Fig. 5C) nearly identical to and coeluting with the peaks in the m/z = 414 to 217

fragmentogram of the oil. No attempt was made to rigorously characterize the two major, early eluting peaks in the oil (Fig. 5A). They are tentatively assigned by their GC retention times and relative abundances compared to the known C_{29}-steranes as (20*S*) and (20*R*), 24-*n*-propyl-13β,17α(H)-diacholestanes (**2fA** and **2gA**). The isomerizate of 24-isopropyl-5β-cholestane (**1bD**) was prepared and analyzed in the same way (Fig. 5D), but its components could not be confirmed with certainty in the geological lipids (Fig. 5, A and E). The m/z = 414 to 217 fragmentogram of an immature rock extract (Fig. 5E) shows mainly the 5α, 14α, 17α(H), 20*R* component, **1aA**, because the rock is not very thermally mature. A smaller abundance of 24-*n*-propyl-5β-cholestane (**1bA**) was identified in this sample by GCMS coelution with an authentic standard.

High concentrations of the rare C_{30}-sterol (24*E*)-24-propylidenecholesterol (**3B**) have been found in three species of unicellular and filamentous algae: *Sarcinochrysis marina* (*16*), *Nematochrysopsis roscoffensis* (*17*), and a closely related but still unidentified sarcinochrysidalean alga (*18*). Minor amounts of 24-*n*-propylcholesterol (**3gA**) were also found. The two named algal forms have been assigned to the order Sarcinochrysidales, originally assigned to the Chrysophyceae (golden brown algae) (*19*). All members now classified as Sarcinochrysidales are marine; most are benthic (for example, *N. roscoffensis*), although *S. marina* is planktonic. Gayral and Billard (*20*) noted morphological features of the Sarcinochrysidales that have "not yet [evolved into] those of authentic Phaeophyceae" (brown algae).

Also, compounds they considered to be diagnostic of the Phaeophyceae are lacking in the Sarcinochrysidales, such as alginic acid in the cell walls and fucosterol (**3I**), the major sterol of brown algae. However, the stereochemistries of the double bond in the (24*E*)-24-propylidenecholesterol (**3B**) from *N. roscoffensis* and in fucosterol (**3I**) are similar. This similarly supports a "possible relationship between the Sarcinochrysidales and the Phaeophyceae (*17*)" or suggests that the order Sarcinochrysidales is phylogeneti-

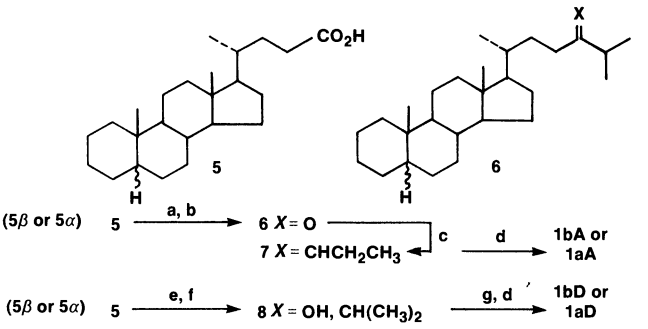

Fig. 2. Schemes for syntheses of C_{30}-steranes: a, $SOCl_2$; b, i-PrMgBr, VCl_3, or $(i\text{-}Pr)_2CuMgBr$; c, NaH, $[Ph_3PCH_2CH_2CH_3]Br$; d, H_2, Pd/C; e, H_3PO_4, CH_3OH; f, i-PrMgBr; g, $POCl_3$, pyridine.

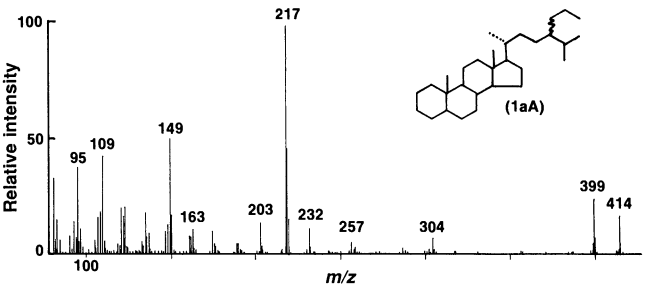

Fig. 3. Mass spectrum of 24-*n*-propylcholestane, obtained from a VG Micromass 7070H mass spectrometer at 70 eV interfaced with an HP 7620A gas chromatograph. Samples were introduced through a 60-m DB-1 fused silica capillary column with H_2 carrier gas; temperature program, 150° to 310°C at 2°C per min.

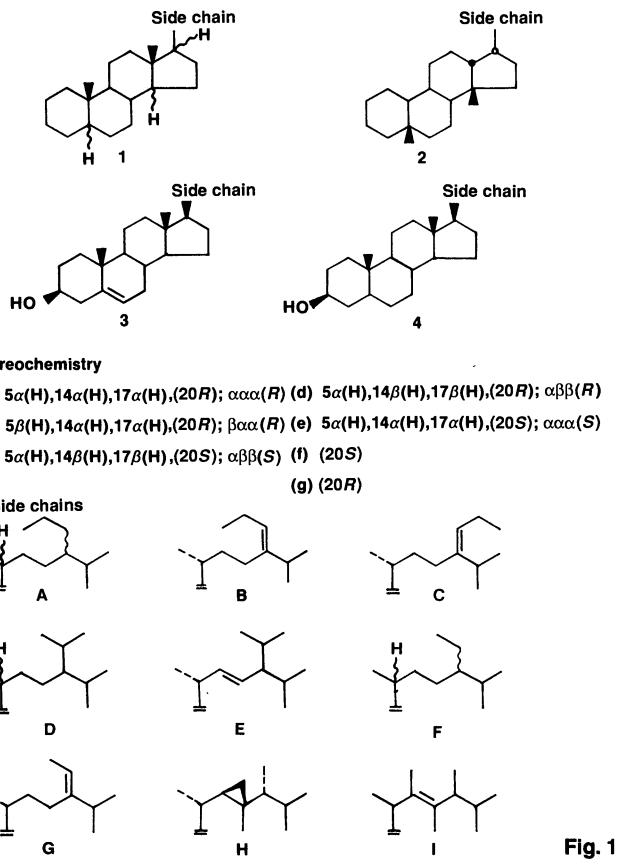

Fig. 1. Structures of selected steroids.

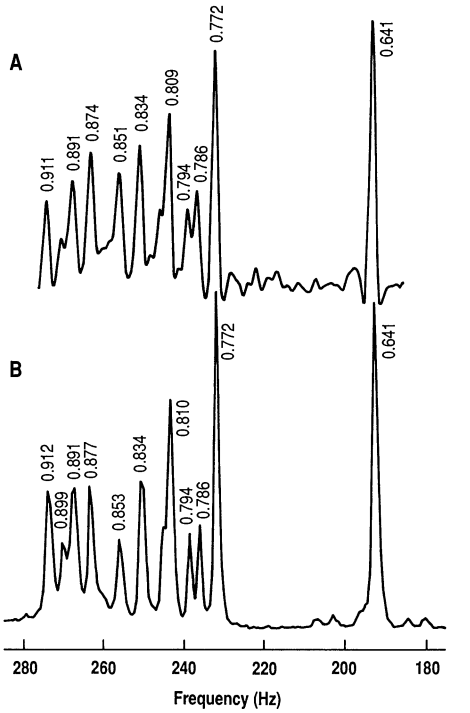

Fig. 4. Region of methyl resonances in 300-MHz H NMR spectra from (**A**) isolated C_{30}-sterane and (**B**) 24-*n*-propylcholestane; the spectra are identical.

cally between the Chrysophyceae and the Phaeophyceae (*20, 21*).

Trace amounts of 24-propylidene sterols (**3B** and **3C**) also have been found in marine invertebrates (*22*) and are most likely of dietary origin. Grazing organisms, particularly in their zooplanktic cycle, may provide fecal pellets that enrich sediments in C_{30}-sterols and sterenes that ultimately yield C_{30}-steranes when thermally mature (*23*).

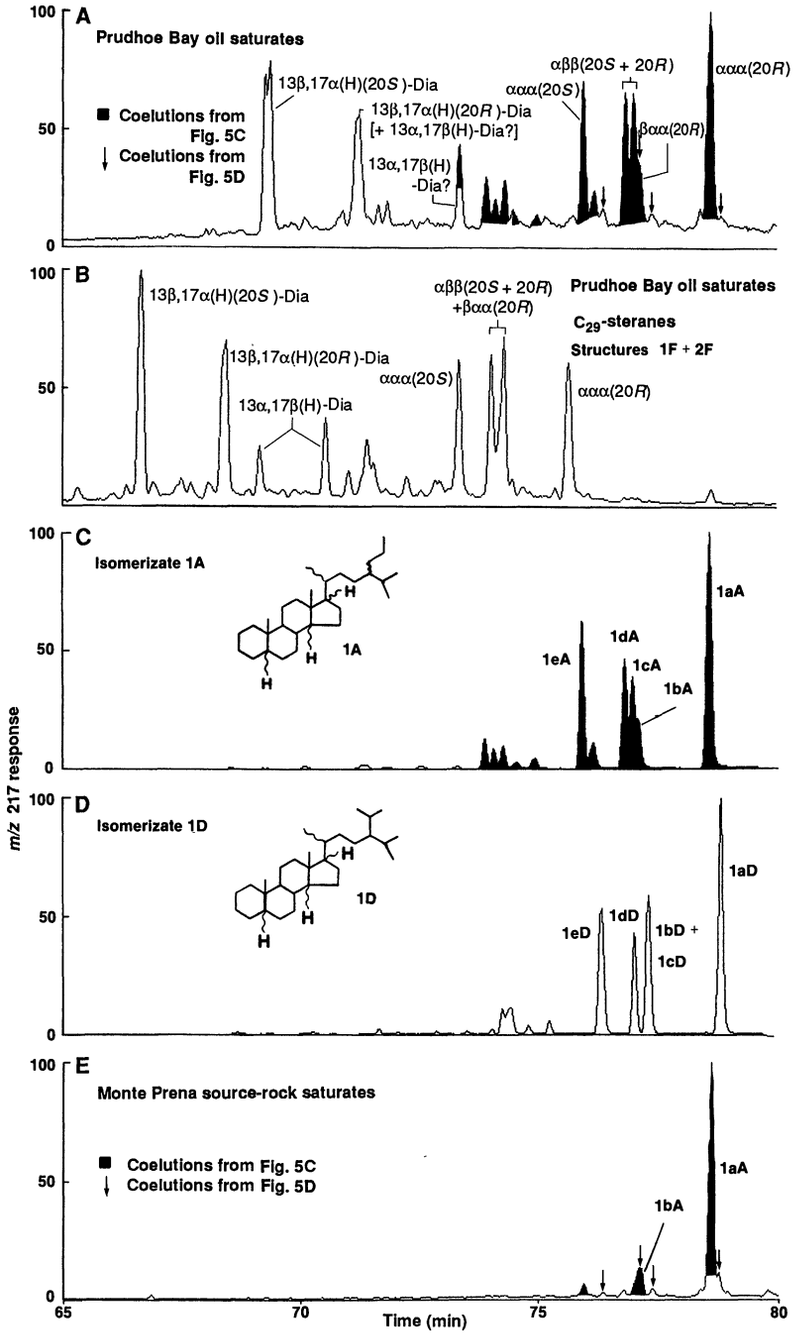

Fig. 5. Comparisons of C_{30}-sterane distributions. (**A**) Saturates, Prudhoe Bay oil (Sadlerochit Formation, 8920 to 9048 feet), metastable reaction monitoring (MRM) m/z = 414 to 217 (C_{30}-steranes) with indications of positions of coeluting peaks from coinjections of catalytic dehydrogenate isomerizate (CDI) mixtures shown in (C) and (D). Conditions as for Fig. 3. (**B**) Saturates, Prudhoe Bay oil, MRM m/z = 400 to 217 (C_{29}-steranes) for comparison of retention times in (A). The C_{29}-sterane ααα20*R* isomer was isolated from the oil and proven by comparison with standards to be a mixture of 24*S* and 24*R*, 24-ethylcholestanes. (**C**) CDI products prepared by heating **1bA** with Pd/C in an evacuated sealed tube at 260°C for 72 hours. MRM m/z = 414 to 217. Stereochemical assignments were based on relative retention times compared with previously assigned cholestane epimers (*14*) except for synthetic isomers reported in this work. (**D**) CDI products prepared by heating **1bD** with Pd/C in an evacuated sealed tube at 260°C for 72 hours. MRM m/z = 414 to 217. Stereochemical assignments as in (C). (**E**) Saturates, Monte Prena, Italy, outcrop (immature) source rock bitumen, GCMS 697. Labeled peaks indicate compounds identified by coinjection of synthetic standards.

Although (4-desmethyl) C_{30}-sterols have been suggested as reliable indicators for marine organisms (*24*), recent work shows that all C_{30}-sterols are not marine specific. Wünsche *et al.* (*5*) reported gorgosterol (**3H**), gorgostanol (**4H**), and 22-dehydro-24-isopropylcholestanol (**4E**) in freshwater (nonmarine) lacustrine sediments; therefore, nonmarine precursor organisms are implied. Also, Robinson *et al.* (*6*) tentatively identified 22,23,24-trimethyl-5α(H)-cholest-22-en-3β-ol (**4I**) in sediments from a saline lake. This work, however, shows that a structural distinction between these nonmarine C_{30}-sterols and the C_{30}-steranes, which are 24-*n*-propylcholestanes (**1A**), is prevalent in marine oils and source rocks (*1, 2*).

A possible exception (*25*) is that Chinese salt lake sedimentary rocks from Bohai Basin apparently contain C_{30}-steranes. We examined one of these rocks (W9, 3144 to 3145 m, Oligocene) and confirmed the presence of 24-*n*-propylcholestanes; amorphous kerogen was found by microscopic examination, and such kerogen is consistent with sedimentation in a saline lake. However, "small amounts of marine fossils" have been identified (*26*) in some Bohai Basin sediments, and they indicate that remnant marine organisms may have been present during deposition of our sample.

The 24-*n*-propylcholestanes (**1A**) have been identified in sedimentary rocks and petroleum dating back to the Devonian (~360 × 10^6 years), but they have not been found in Cambrian and Precambrian petroleum (*2, 27*), because they were either absent or too dilute to identify. We have studied samples of the Alum shale, Sweden, which range from Middle Cambrian to Lower Ordovician in age (*28*). Although ample C_{26}- to C_{29}-steranes were present, 24-*n*-propylcholestanes could not be detected in any of these marine rocks. This incomplete survey suggests that these C_{30}-steranes appeared in the early Paleozoic and, by implication, that the algae that biosynthesize their precursor sterols emerged between the Early Ordovician and the Devonian.

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9. The starting materials for these syntheses are commercially available 5β-cholanic acid and 5α-cholanic acid (*10*), respectively. The synthetic routes to the

steranes involved the coupling of 5β-cholanyl chloride with either bromomagnesium diisopropylcuprate [$(i\text{-}Pr)_2CuMgBr$] or isopropylmagnesium bromide (i-PrMgBr) in the presence of vanadium trichloride (*11*) to produce the ketones **6**. The Wittig reaction of **6** with the ylid derived from *n*-propyl triphenylphosphonium bromide [$(Ph_3PCH_2CH_2CH_3)Br$] furnished the olefins (*7*) as an *E-Z* mixture. Hydrogenation of these olefins with 10% Pd on C provided the desired alkanes (**1bA** and **1aA**) as a diastereomeric mixture at C-24. Significant impurities (3 to 5%) of 5β and 5α-24-methylcholestane were detected by GCMS analysis of the 5β and 5α-24-*n*-propylcholestane products, respectively, which were generated in a retroene reaction during the catalytic hydrogenation step. {A retroene process is the reverse of the "ene synthesis" [W. Oppolzer and V. Snieckus, *Angew. Chem. Int. Ed. Engl.* **17**, 476 (1978)] in which a hydrocarbon RH formally adds to an olefin.} They were removed by preparative, nonaqueous, reversed-phase HPLC. No attempt was made to resolve 24*R* and 24*S* isomers because the isolated C_{30}-sterane should be a mixture of both isomers. Epimerization of the C-24 position occurs early in sediment diagenesis (*12*). The synthetic route to the isopropylcholestanes (**1bD** and **1aD**) involved addition of i-PrMgBr to 5β or 5α methyl cholanate to produce principally the tertiary alcohol **8**. Dehydration of **8** with phosphorus oxychloride led to a mixture of olefins that was hydrogenated at high pressure to give impure samples of **1bD** and **1aD**. The principal difficulty with this route was that the competitive reduction of the intermediate ketone **6** by i-PrMgBr led to a small but significant amount of a secondary alcohol that cochromatographed with **8**. This contaminant led ultimately to 5β and 5α-cholestane, which were removed by HPLC.

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Effects of Fish and Plankton on Lake Temperature and Mixing Depth

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A comparative study of small temperate lakes (<20 square kilometers) indicates that the mixing depth or epilimnion is directly related to light penetration measured as Secchi depth. Clearer lakes have deeper mixing depths. This relation is the result of greater penetration of incident solar radiation in lakes and enclosures with high water clarity. Data show that light penetration is largely a function of size distribution and biomass of algae as indicated by a relation between the index of plankton size distribution (slope) and Secchi depth. Larger or steeper slopes (indicative of communities dominated by small plankton) are associated with shallower Secchi depth. In lakes with high abundances of planktivorous fish, water clarity or light penetration is reduced because large zooplankton, which feed on small algae, are reduced by fish predation. The net effect is a shallower mixing depth, lower metalimnetic temperature and lower heat content in the water column. Consequently, the biomass and size distribution of plankton can change the thermal structure and heat content of small lakes by modifying light penetration.

THERMAL STRATIFICATION OF THE water column occurs in most temperate lakes during late spring and summer (*1*). The epilimnion or upper mixed layer, with a fairly uniform temperature, is separated from the cooler bottom layer or hypolimnion by a middle layer or metalimnion where temperature drops rapidly with depth. The mixing depth of a lake has a fundamental influence on the physical, chemical, and biological processes that take place in the water column (*2–5*).

Wind-induced transport of heat to deeper strata has been considered more important in determining the mixing depth than the direct absorption of solar energy (*3, 6*). In this report, we show that water clarity, associated with the biomass and size distribution of plankton (algae and zooplankton), can influence the mixing depth and heat content of small lakes. Physical characteristics of lakes, such as light and temperature, have a profound influence on their biology, but the effect of organisms on lake physics has been uncertain and generally thought to be of secondary importance.

Solar irradiance may be reflected at the surface, backscattered by planktonic particles to the atmosphere, and absorbed as heat by water. The balance between the two losses and heat absorption determines the heat content of the surface mixed layer. Secchi depth, an index of light penetration, can be as shallow as 0.5 m or less in hypereutrophic and highly colored (brown-water) lakes (*7*) or as deep as 20 m in clear lakes, such as Lake Tahoe (*8*). The deeper the Secchi depth, the greater the absorption of solar radiation by the water, and the smaller the loss of solar energy to the atmosphere (*9*).

Algae have a major influence on water clarity and attenuation of light in noncolored lakes (*7, 10*) and in oceans (*11*). Light is both scattered and absorbed by algae, and the effects depend on their size distribution and biomass. Smaller algae (pico- and nanoplankton, 0.2 to 20 μm) have greater absorption and scattering per unit mass than larger algae (>20 μm) because their surface area per unit mass is larger (*12*). Fish predation on large zooplankton, which graze mostly on small algae, is a strong determinant of the biomass and size distribution of algae (*13*). This effect has been supported by many experimental and empirical studies where low abundance of planktivorous fish was associated with high abundance of large zooplankton, low biomass of algae, and improved water clarity (*13–15*). Therefore fish predation, and the resulting size distribution and biomass of plankton, might influence thermal structure of small lakes by changing water clarity where wind-induced transport of solar heating to deeper water is less important.

We performed a series of manipulation experiments in large enclosures situated in Lake St. George, Toronto, Ontario (*16*). During May through August, 1986 and 1987, we collected data on the size distribution and biomass of the entire plankton community, from bacteria to zooplankton, measured as particulate phosphorus (*PP*) (*17*). We also measured size distribution and abundance of *Daphnia*, algal biomass (chlo-

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