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ORGANIC GEOCHEMICAL ANALYSIS OF LATE GLACIAL AND EARLY HOLOCENE ECOSYSTEM CHANGES: A CASE STUDY FROM NORTHERN NEW ENGLAND LAKES



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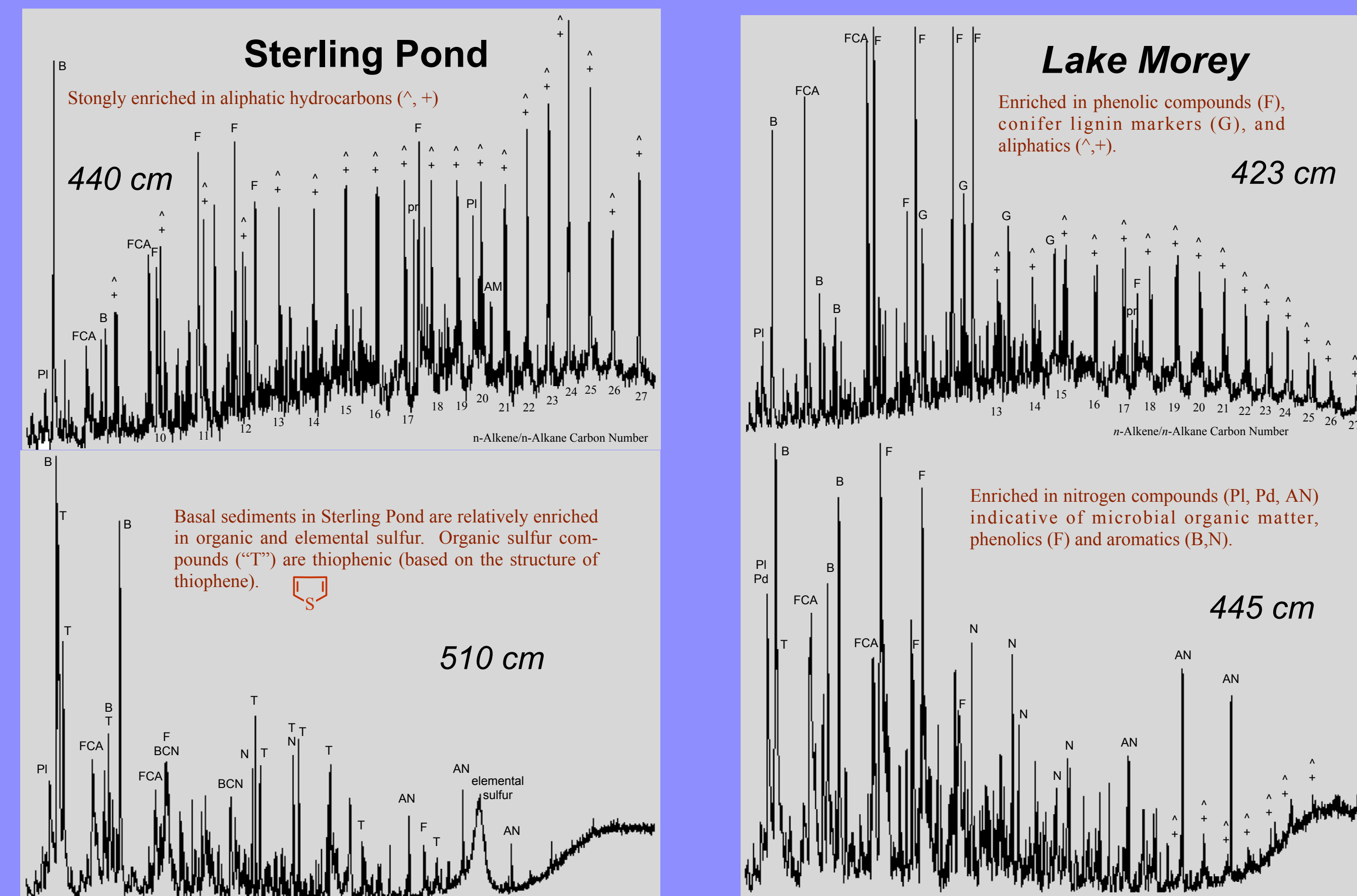


ABSTRACT

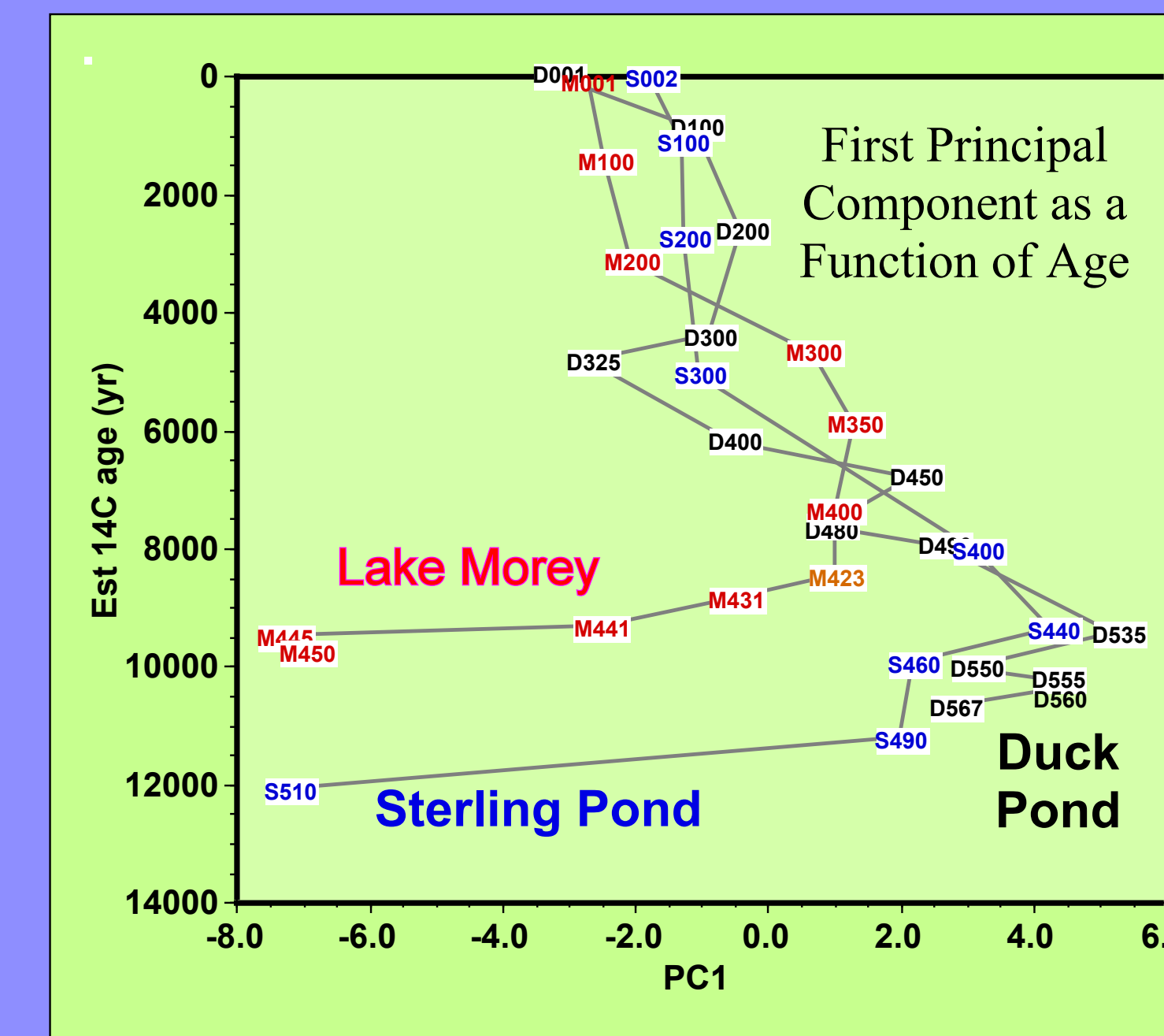
The lacustrine sedimentary archive of organic remains provides important evidence for the reconstruction of the environmental histories of lakes and their watersheds, recording the response of the Earth's biota to changes on scales varying from the local to the global. The last Glacial-Interglacial transition presents an opportunity to investigate how, and at what rates, watershed and lake ecosystems were established on once glaciated, carbon and nutrient-poor landscapes. The small lakes of northern Vermont (USA) provide an appropriate setting in which to investigate such changes.

As part of a multidisciplinary study of three Vermont lake sediment cores, samples ranging in age from approximately 0 to 13 ka were subjected to molecular organic geochemical analysis by pyrolysis-gas chromatography/mass spectrometry (Py-GC/MS). Diagnostic compounds in the pyrolyzates (e.g., indole and guaiacol) permit discrimination between the principal organic matter sources (e.g., algal/bacterial and higher plant) and their shifting proportions over time as a function of environmental change. While the nature of this shift is similar, it does not occur at the same rate in all three lakes, indicating the profound influence of local conditions.

Pyrolysis-Gas Chromatography/Mass Spectrometry Total Ion Current Pyrograms

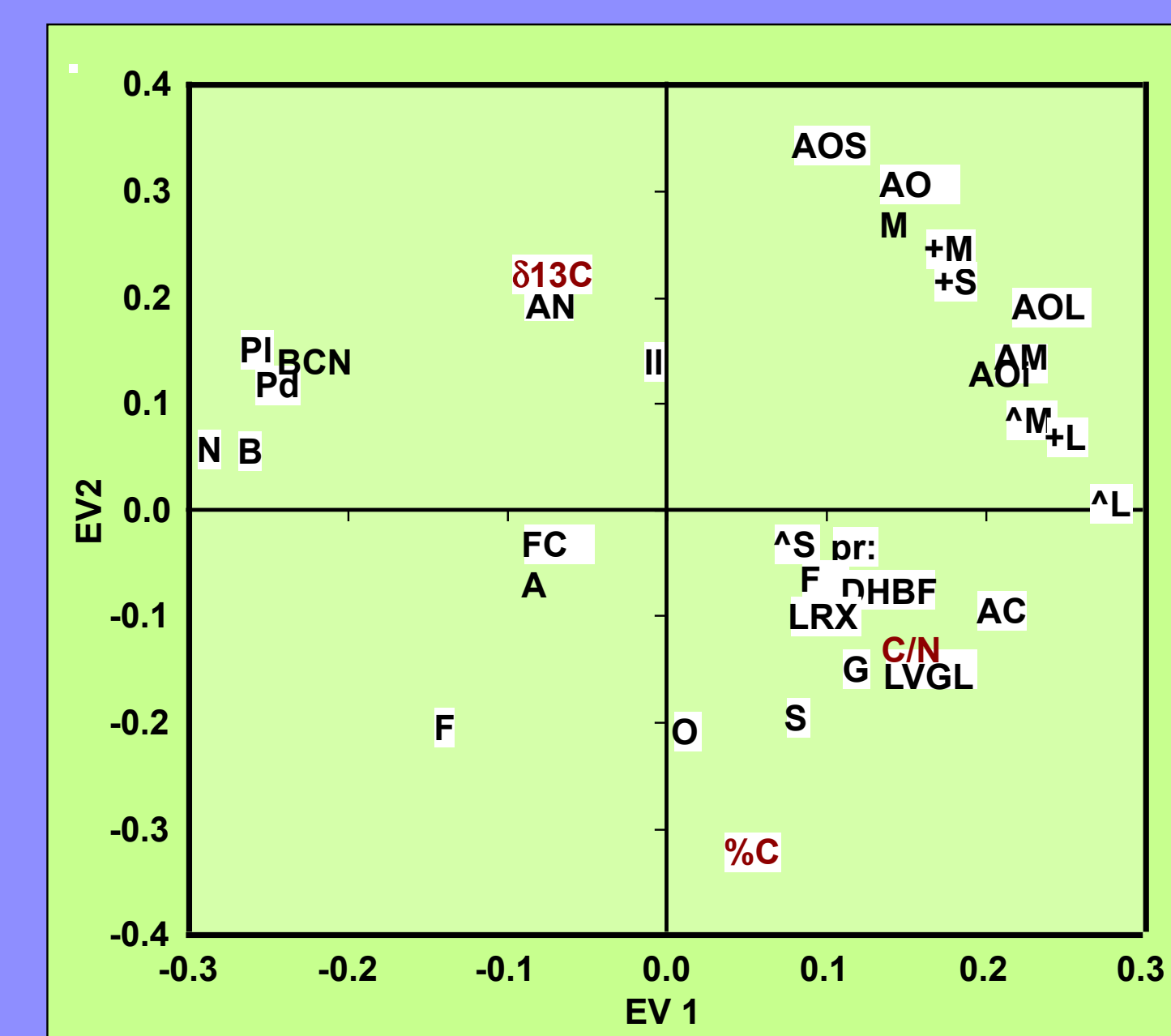
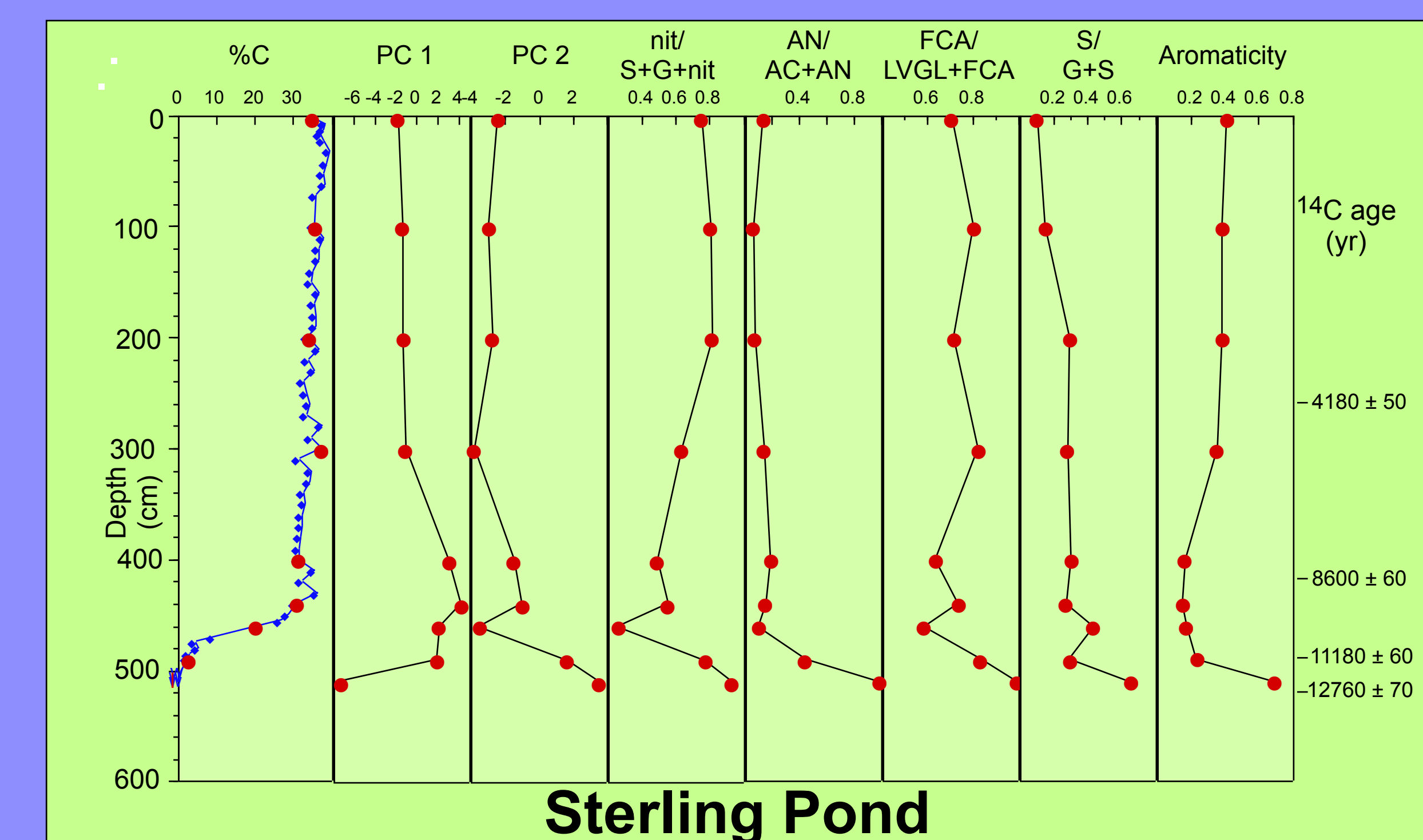
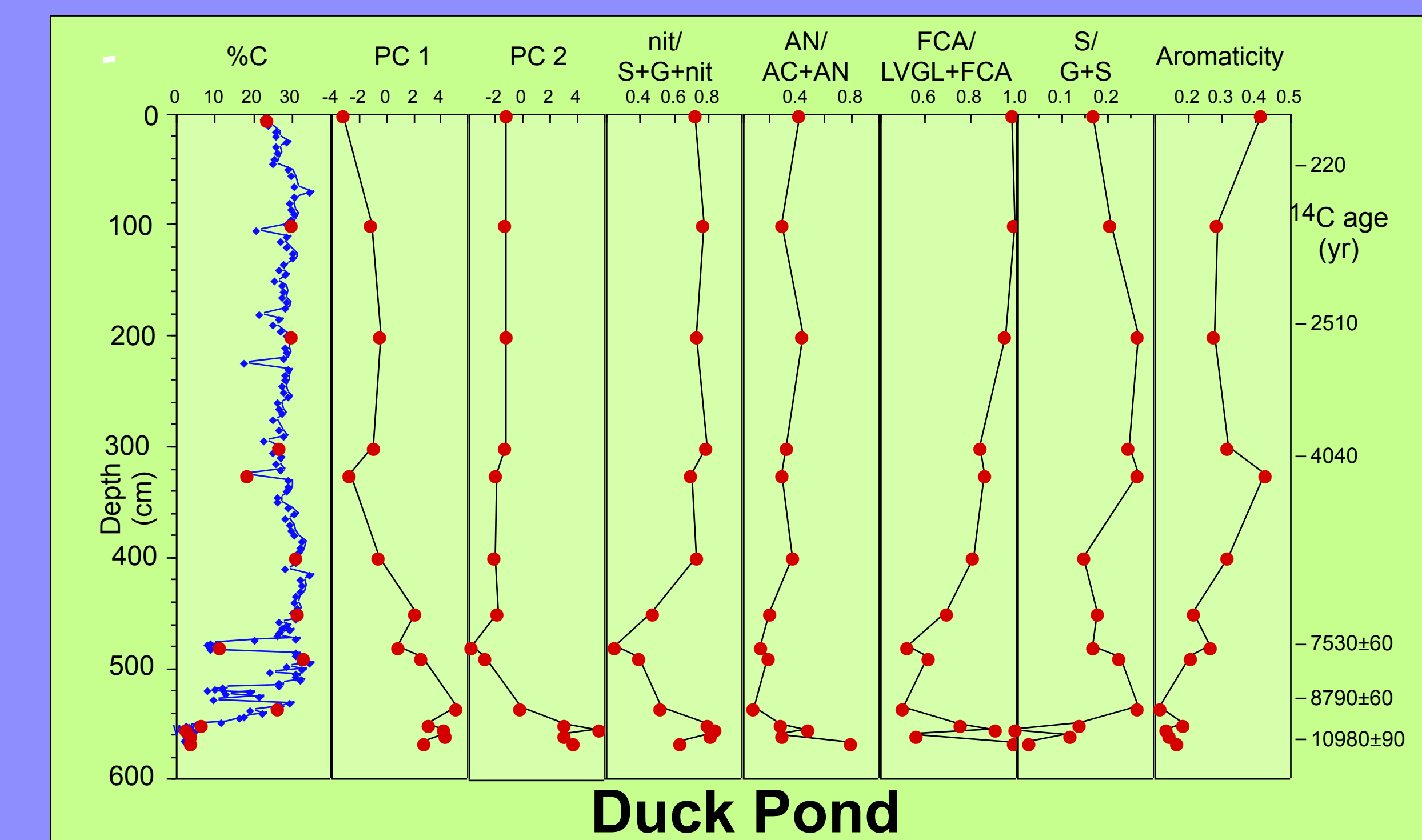
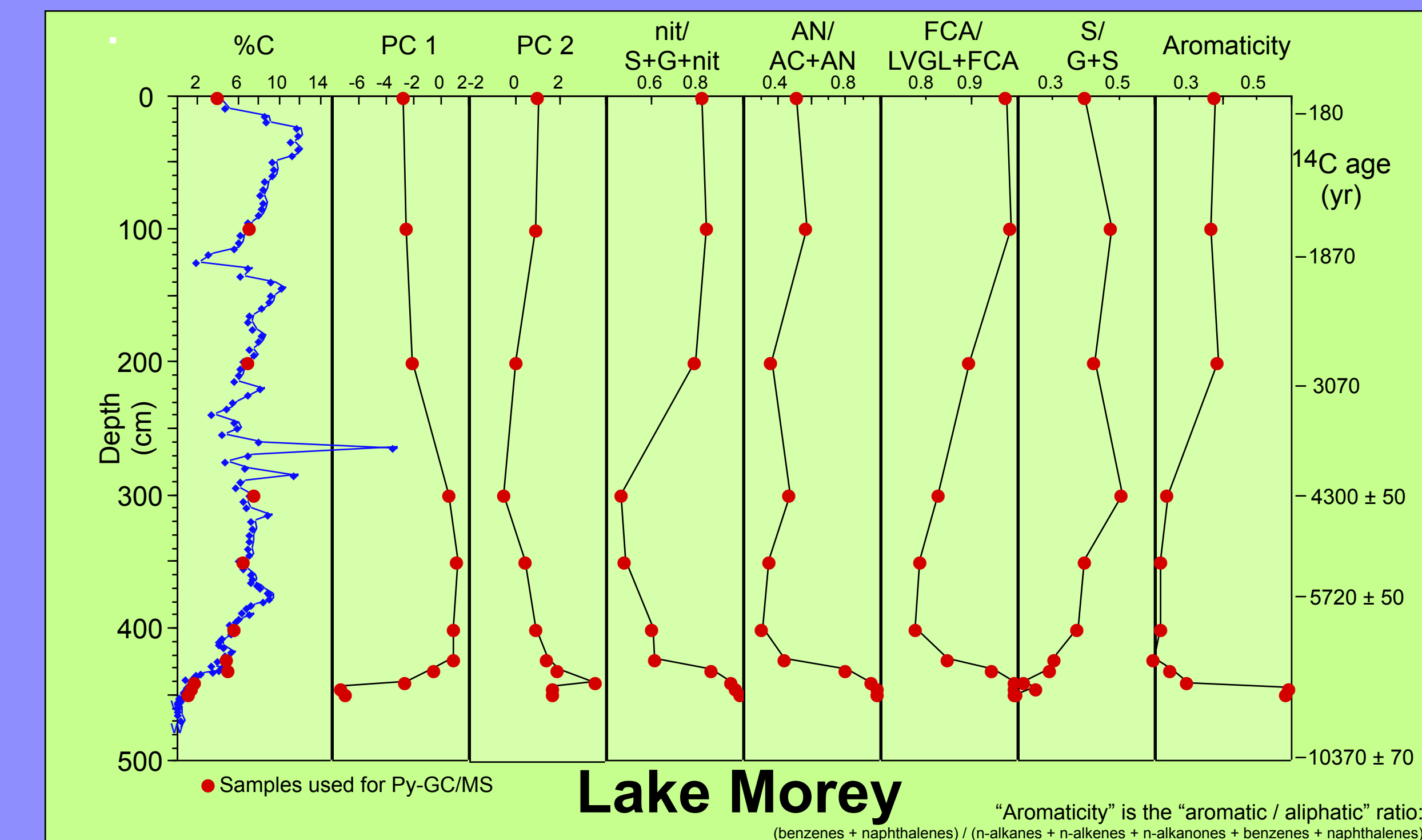


Examples of pyrolyzates from the post-glacial transition at the base of the sediment sequences in two lakes. Note the relative increase in aliphatic hydrocarbons (^, +) and lignin markers (G) across the transition, as well as the relative decrease in nitrogen compounds (PI, Pd, BCN, AN). The relative sulfur enrichment seen in the pyrolyzate of the 510 cm Sterling Pond sample might be the result of early diagenetic (microbial) processes in the presence of sulfate-rich pore waters, the source of which, however, is not known to the authors.



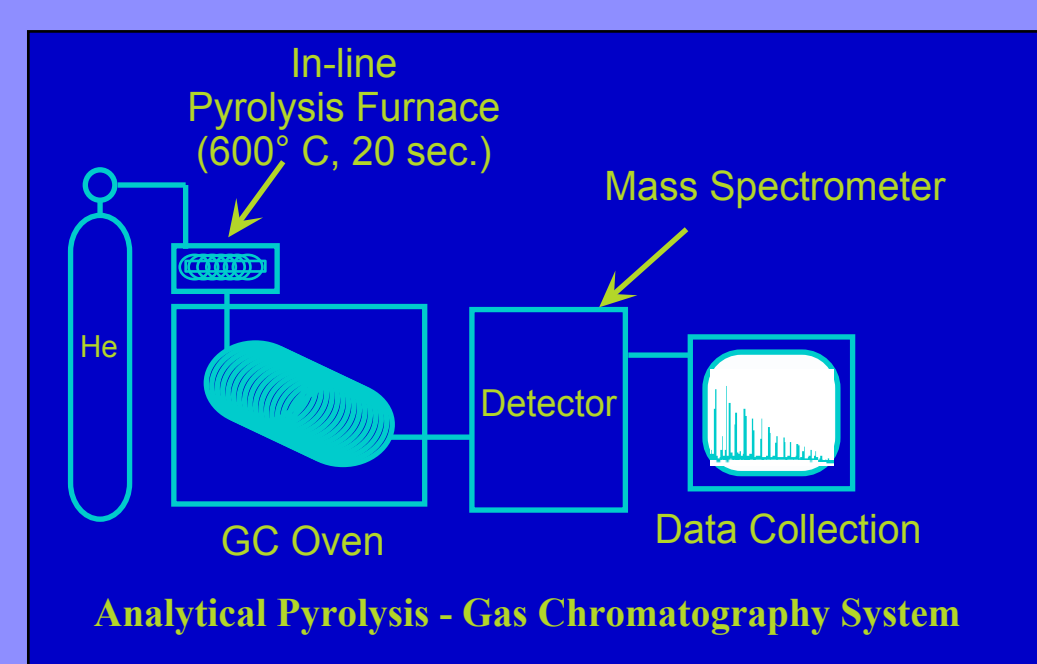
The first principal component deflects strongly at the base of the sedimentary sequence in the Lake Morey and Sterling Pond cores (above), reflecting significant organic matter changes at the last Glacial-Interglacial transition. (The details on the molecular level may be seen in the pyrograms on the left and in summary form in the depth plots on the right.) The transition at Duck Pond is chemically different, showing a strong deflection in the second principal component (see figure at right), corresponding to a change from aliphatic OM to a predominance of lignin-derived material.

The timing of the transition also varies, being earliest at the more northerly, higher elevation Sterling Pond site and latest at the more southerly, lower elevation Lake Morey site. The bulk geochemical parameters (see poster SS3.09.131) organic carbon content, C/N and $\delta^{13}C$ show a similar effect.



The eigenvectors ("EV") for the first two principal components show the spatial relationships between the 29 compound groups, as well as organic carbon, C/N and $\delta^{13}C$. The nitrogen compounds (PI, Pd, BCN, AN) group together in the upper left quadrant, while the lignin markers (G, S) and higher plant carbohydrate markers (LVGL, LRX) group together in the lower right, along with %C and C/N. Long chain (aliphatic) compounds group in the upper right.

Most of the parameters plotted above (as a function of depth) show a strong deflection at the base of the sequence, corresponding to the post-glacial transition. The OM shifted from algal/bacterial at the very base to terrestrial, with a (different) algal/bacterial assemblage tending to regain dominance in the younger sediments. This is shown clearly in the ratio of nitrogen compounds to lignin markers (nit/S+G+nit) and in the ratio of carbohydrate markers (FCA/LVGL+FCA). Although the cores show similar general trends, they are in fact chemically different from one another, reflecting different OM inputs and local environmental conditions.

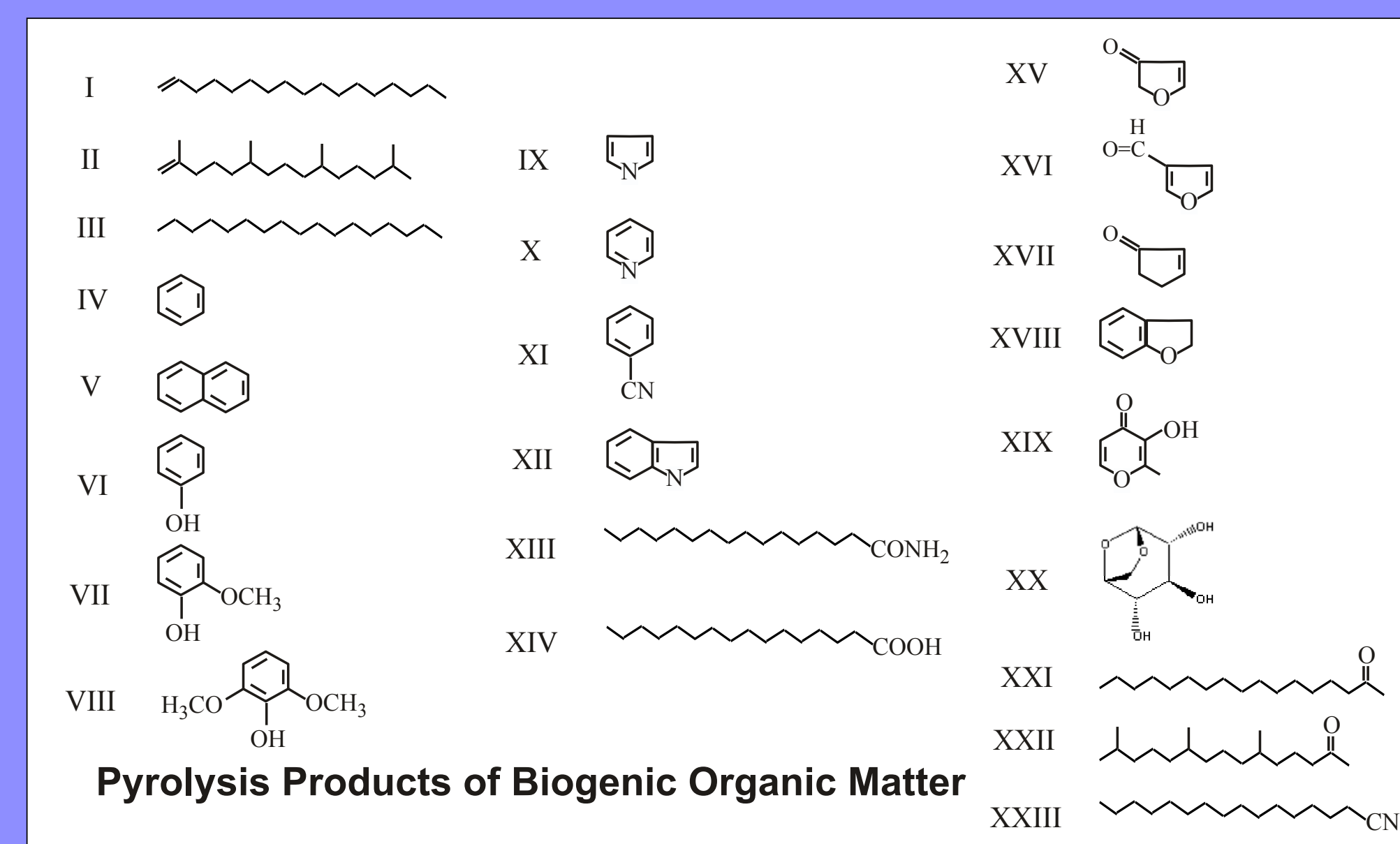


METHODS

Cores from three northern Vermont lakes (Lake Morey, Duck Pond, Sterling Pond - see poster SS3.09.131) were subsampled. Pyrolysis-gas chromatography/mass spectrometry (Py-GC/MS) was performed using a CDS 120 pyroprobe, coupled to a HP 5890 gas chromatograph with a HP 5970 mass selective detector and a 50 m J&W Scientific DB-5MS column (0.2mm i.d., film thickness 0.33 mm). A measured amount (a few mg) of dry, powdered sample was pyrolyzed in a flow of helium for 20 sec. in a platinum coil at 600 °C, as measured by a thermocouple in the sample holder. The GC oven was operated under the following program: isothermal for 5 min at 40 °C; temperature programmed at 5 °C/min. to 300 °C and then isothermal for 30 min. The MS was operated in full scan (50-450 Da, 1.21 scans/sec., 70eV ionization voltage).

Quantification of data was accomplished using Hewlett Packard GC/MS software G1701AA (version A.03.00). All data was adjusted by response factors previously determined for each compound using this GC/MS instrument and normalized to the sum of all peaks in each sample.

Multivariate analysis was performed using JMP version 3.1.5 software (SAS Institute Inc, 1995). The 135 compounds from Py-GC/MS data set (corrected and normalized as described above) were grouped into 29 categories according to compound class and summed, as suggested by a preliminary analysis using the individual compounds. A correlation matrix was computed on the 29 categories, along with total organic carbon, the C/N ratio and $\delta^{13}C$, followed by a principal components analysis.



Pyrolysis Products of Biogenic Organic Matter

^S	C ₇ -C ₁₃ n-alk-1-enes	I	indoles	XII
^M	C ₁₄ -C ₂₀ n-alk-1-enes	I	n-alkylamides	XIII
^L	C ₂₁ -C ₂₉ n-alk-1-enes	I	n-alkanoic acids	XIV
pr:	priest-1-ene	II	furanones	XV
+S	C ₇ -C ₁₃ n-alkanes	III	furancarboxaldehydes	XVI
+M	C ₁₄ -C ₂₀ n-alkanes	III	cyclopentenones	XVII
+L	C ₂₁ -C ₃₁ n-alkanes	III	DHBF	XVIII
B	benzenes	IV	LRX	XIX
N	naphthalenes	V	LVGL	XX
F	phenols	VI	AOS	XXI
G	guaiacols	VIII	AOM	XXI
S	syringols	VIII	AOL	XXI
PI	pyrroles	IX	AOi	XXII
Pd	pyridines	X	AN	XXIII
BCN	benzonitriles	XI		