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Radiocarbon chronology of Black Sea sediments

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Abstract—Accelerator Mass Spectrometer (AMS) radiocarbon analyses have been made on 102 samples from 12 sediment cores and 23 samples from two water column profiles. These materials, collected during the first leg of the 1988 joint U.S.–Turkish Black Sea Expedition, provide the most comprehensive radiocarbon chronology of Black Sea sediments yet attempted. Radiocarbon analyses from carefully collected box cores and a mollusc shell collected live in 1931 suggest the pre-bomb surface waters had a $\Delta^{14}C$ value of −55‰ (460 years) and that the maximum detrital correction for radiocarbon ages of Unit I sediments is 580 years for the organic carbon and 260 years for the carbonate fractions. Evidence does not support the 1430–2000 year pre-bomb surface water and/or detrital corrections argued for in past studies. The best estimates for the age of the beginning of the final invasion of the coccolithophore *Emiliania huxleyi* (Unit 1/2 boundary of Ross and DEGENS, 1974, *The Black Sea—geology, chemistry and biology*, pp. 183–199) and the age of the first invasion of *E. huxleyi* (Unit I/II boundary of HAY *et al.*, 1991, *Deep-Sea Research*, **38**, S1211–S1235) are 1635 ± 60 and 2720 ± 160 years BP, respectively. Sapropel formation began at approximately 7540 ± 130 years BP at all depths in the basin, a pattern in disagreement with those predicted by existing time-evolution models of sapropel formation for this basin. Our data suggest that the oxic–anoxic interface has remained relatively stable throughout the Holocene, is controlled largely by the physical oceanography of the basin, and has not evolved as assumed by previous workers.

INTRODUCTION

THE Black Sea (Fig. 1) is the world's largest anoxic basin in both areas (508,000 km^2) and volume (536,000 km^3) (MURRAY, 1991), with oxygen-free conditions in water below approximately 100–150 m depth. A permanent halocline effectively isolates the oxygenated near surface waters, with an average salinity of 18 ppt from the anoxic deeper waters, with an average salinity of 22 ppt. This sharp halocline and associated chemocline results from an inflow, through the Bosporus Strait, of saline (35 ppt) Mediterranean waters into the bottom of the basin, while the low salinity of surface water is maintained by an excess of combined river inflow and precipitation over evaporation. The anoxic condition of the basin largely determines the chemistry and biology of the water column as well as the character of the sediments preserved at the sea bottom.

The unique conditions found in the Black Sea have held the interest of the scientific community for much of the past century. The first major deep-sea expedition to the Black Sea was made by Andrusov, which also recovered the first anoxic sediments from the basin

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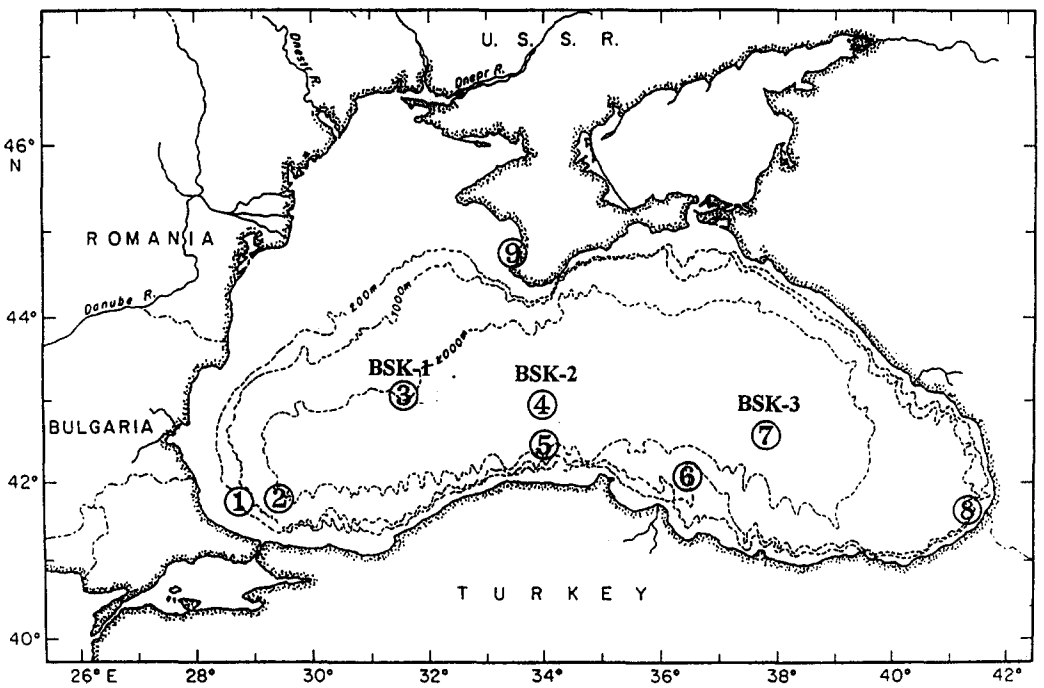

Fig. 1. Map of the Black Sea and surrounding land masses showing the sample locations for sediment cores, hydrocasts, pre-bomb mollusc and sediment traps. Details relating to each core, hydrocast and sediment trap are given in Table 1.

(ANDRUSOV, 1890). The geological history of the basin was initially studied by ARKHANGELSKI and STRAKHOV (1938), and the formulation of the basic hydrography was first addressed by ANTIPA (1941) and NEUMANN (1942).

The major U.S. oceanographic expeditions conducted in this basin prior to recent efforts were aboard the vessel R.V. *Atlantis* II in 1969 and the R.V. *Chain* in 1975. Most recently, the United States National Science Foundation sponsored the 1988 Black Sea Expedition: a U.S.–Turkish effort designed to remedy deficiencies in previous datasets resulting from either new scientific insights, improved analytical capabilities, and/or improved spatial coverage (MURRAY, 1991). Several comprehensive reviews of Black Sea studies resulting from these, and other, oceanographic expeditions have been published (e.g. VODIANITSKII, 1968; FILIPPOV, 1968; DEGENS and ROSS, 1974; SKOPINTZEV, 1975; SOROKIN, 1983; MURRAY, 1991; IZDAR and MURRAY, 1991).

Despite extensive study and several compilations of the results of the major oceanographic expeditions to this basin, there remain unresolved problems, one of which is the establishment of a firm chronology for the Late Quaternary sediments preserved in this basin (ROSS and DEGENS, 1974; DEGENS *et al.*, 1980; CALVERT, 1987; HAY *et al.*, 1991; CALVERT *et al.*, 1991; MURRAY *et al.*, 1991; CRUSIUS and ANDERSON, 1992). The disagreement regarding the chronology of the late Quaternary sediments of the Black Sea centers around the disparity in the ages obtained from radiocarbon dating of the sediments versus

the counting of laminae found in the sediments which have been interpreted as annual couplets or varves. Previously, the varve chronology has been used to suggest that the first occurrence of *Emiliania huxleyi* in the basin occurred at approximately 1000 years BP (DEGENS *et al.*, 1980) to 1700 years BP (ARTHUR *et al.*, in press), whereas previous radiocarbon analyses suggested the first appearance of *E. huxleyi* at approximately 3100 years BP (ROSS and DEGENS, 1974; CALVERT, 1987). Published radiocarbon results also suggest that the transition from lacustrine lutites to finely-laminated sapropellic muds occurred at approximately 7090 years BP vs an estimated 5080 years BP from varve counting.

Determining the correct chronology of Black Sea sediment has major implications for understanding the history of the development of anoxia in the basin, the flux and preservation of organic matter in the basin, interpretation of carbonaceous shale formation, the relationship between environment and the development of the Black Sea from a lacustrine to an anoxic basin and consequently, the role played in the development of this region's human history.

This paper reports on the results of 102 AMS radiocarbon measurements made on sediments from 12 cores, 23 dissolved inorganic carbon (DIC) water column measurements from two profiles, and two sediment trap samples of particulate organic matter (POM). All material was recovered in May 1988 during Leg 1 of R.V. *Knorr* cruise 134, stored, and sampled for the express purpose of being AMS radiocarbon dated. This material is of much improved quality compared with material from cores collected during the 1969 and 1975 U.S. expeditions which were radiocarbon dated using conventional (ROSS and DEGENS, 1974) or AMS methods (CALVERT, 1987). In addition, a mollusc shell collected live in 1931 has been analysed to determine directly the radiocarbon age of the pre-bomb surface waters.

METHODS

Stratigraphy

ROSS and DEGENS (1974) presented the first widely referenced stratigraphy for the Holocene sediments of the Black Sea (Fig. 2). These authors divided the sediments into three lithostratigraphic units. The uppermost unit (Unit 1) is characterized by alternating light and dark microlaminations laid down since the initiation of the most recent invasion of *E. huxleyi*, a coccolithophore that largely comprises the light bands. Unit 2 is a carbonate-poor, fine-grained, organic-rich sapropel containing a brief earlier invasion of *E. huxleyi* in a microlaminated band. Unit 3 consists of alternately light and dark lutites deposited under lacustrine conditions (Fig. 2).

A modification to this scheme was proposed by HAY *et al.* (1991) who reexamined this stratigraphy using material collected during the 1988 U.S.–Turkish expedition. The single difference between the two schemes lies in the placement of the 1/2 boundary (Fig. 2). HAY *et al.* (1991) defined the beginning of the uppermost lithostratigraphic unit as the first occurrence of *E. huxleyi*, which they called the first invasion period. These authors called this Unit I (using Roman numerals to differentiate from ROSS and DEGENS, 1974). Hay *et al.*'s Unit II differs from Ross and Degens' Unit 2 in that the former does not encompass the first brief invasion of *E. huxleyi* nor what they call the transition sapropel deposited above it. The base of Unit II is identical to the base of Unit 2 in ROSS and DEGENS (1974). Unit III is identical to Unit 3 (Fig. 2).

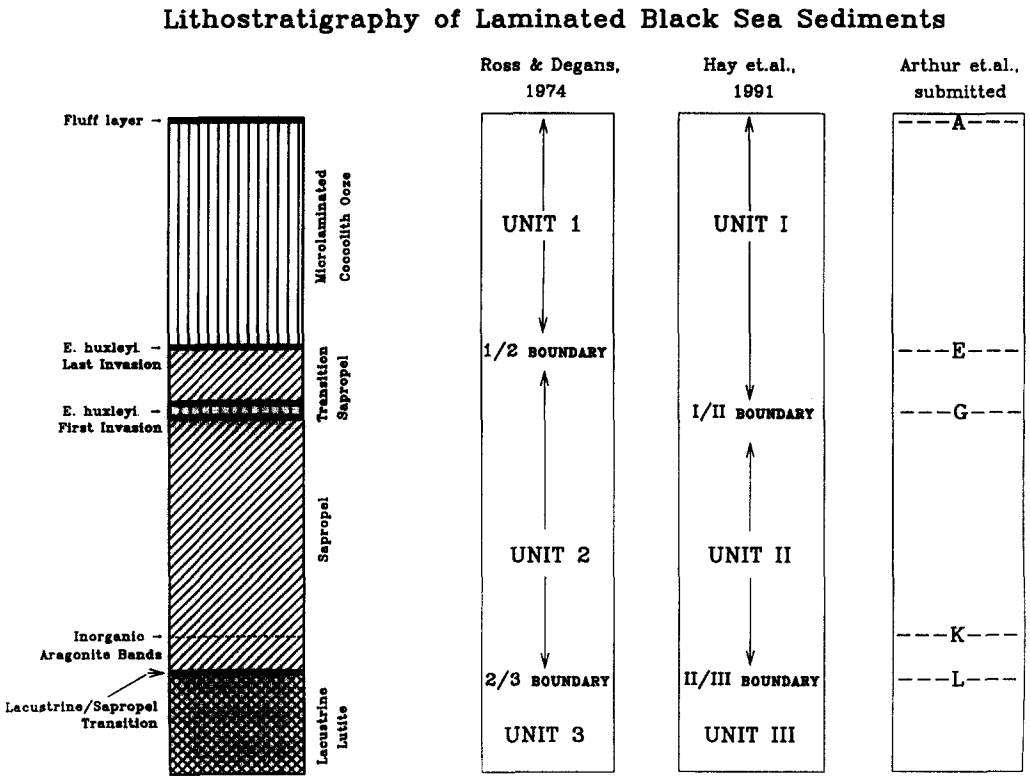

Fig. 2. Comparison of three different lithostratigraphic schemes for Holocene Black Sea sediments (ROSS and DEGENS, 1974; HAY *et al.*, 1991; ARTHUR *et al.*, in press). The scheme of ARTHUR *et al.* (in press) is used throughout this text.

The use of three lithostratigraphic units is insufficient for making detailed correlations among the sediments of the basin. ARTHUR *et al.* (in press) further differentiated these sediments by identifying 12 lithostratigraphic horizons within Units I and II. We mention briefly the five most distinctive and important horizons for the chronology discussed here. The base of the uppermost sediment or fluff (flocculent) layer is defined as Horizon-A. The Unit 1/2 boundary of Ross and Degens is Horizon-E, and the Unit I/II boundary of HAY *et al.* (1991) is Horizon-G. Near the base of Unit II occurs a series of inorganically precipitated aragonite layers first reported in Ross and DEGENS (1974), and the base of these layers is identified as Horizon-K. Finally, the Unit II/III boundary is identified as Horizon-L (Fig. 2).

Radiocarbon

All radiocarbon analyses are reported using the convention of STUIVER and POLACH (1977). Results of seawater and sediment trap sample analyses are reported as $\Delta^{14}C$, or the per mil depletion (−) or enrichment (+) of the sample relative to the oxalic acid standard after the sample and standard radiocarbon activities have been normalized for isotopic fractionation. Sediment samples are reported as conventional radiocarbon ages (years BP)

Table 1. *Sampling locations and water depths for all Box Cores (BC), Giant Gravity Cores (GGC), Hydrocasts (DIC/H_2O) and sediment trap deployments (Sed. Trap) used in this study. Map codes* (1–9) *are used to identify the locations as shown on Fig.* 1. *The type of material analysed for each core or station is listed as Total Organic Carbon and Total Carbonate in the sediments (SED TOC/TCC), Dissolved Inorganic Carbon in seawater* (H_2O *DIC*), *or Total Particulate Organic Matter in the sediment traps (Trap POM/TOC)*

Core/Site	Map code	Latitude	Longitude	Depth (m)	AMS 14C analysis
KNR134-08 BC17	3	42°58'N	31°25'E	2066	SED TOC/TCC
KNR134-08 BC25	4	42°56'N	34°01'E	2217	SED TOC/TCC
KNR134-08 BC55	7	42°45'N	37°35'E	2164	SED TOC/TCC
KNR134-08 GGC01	1	41°53'N	28°49'E	549	SED TOC/TCC
KNR134-08 GGC08	1	41°56'N	28°55'E	897	SED TOC/TCC
KNR134-08 GGC09	2	41°56'N	29°02'E	1259	SED TOC/TCC
KNR134-08 GGC11	2	41°57'N	29°06'E	1490	SED TOC/TCC
KNR134-08 GGC19	3	42°53'N	31°23'E	2096	SED TOC/TCC
KNR134-08 GGC24	5	42°59'N	34°00'E	2195	SED TOC/TCC
KNR134-08 GGC38	7	42°21'N	37°30'E	2030	SED TOC/TCC
KNR134-08 GGC48	8	41°27'N	41°22'E	205	SED TOC/TCC
KNR134-08 GGC65	5	42°18'N	34°00'E	563	SED TOC/TCC
KNR134-08 BSK1	3	43°10'N	31°59'E	2067	H_2O DIC
KNR134-08 BSK2	4	43°00'N	34°01'E	2386	Trap POM/TOC
KNR134-08 BSK3	7	42°23'N	37°35'E	2137	H_2O DIC
M. galloprovincialis	9	44°40'N	33°30'E	<5	Pre-bomb mollusc

where 1950 is the base year and the Libby half-life of 5568 years is used. In addition, reservoir corrections of 460 and 60 years are applied to all marine carbonate and organic carbon ages, respectively. These corrections are based upon the value of the pre-bomb surface water as determined by analysis of a mollusc shell collected live in 1931.

Box cores

Of the 30 box cores collected during R.V. *Knorr* cruise 134, leg 1, one from each of the west, central and eastern sectors of the central Black Sea were selected to provide maximum areal coverage for radiocarbon dating the distinctive Unit I lithostratigraphic horizons across the basin (Table 1 and Fig. 1). The box cores were taken using an Ocean Instruments Mark-III box corer, with recovery dimensions of 50 cm L × 50 cm W × 60 cm H. Due to the high water content of the bottom sediments, modifications were made to the frame of the corer to decrease penetration into the mud. Wooden planks were added to the frame of the corer to increase surface area and the corer was lowered at the slowest winch speed, (approximately 7 m min^{-1}). Even with these modifications, over-penetration occurred in 13 of the 30 cores. None of the three box cores analysed for AMS ^{14}C, however, exhibited any evidence of over-penetration.

A sampling scheme was designed for the placement and ordering of the 10–12 subcores taken from each box core. Two of these subcores were designated for radiocarbon analyses. The first of these subcores (always first to be inserted and removed) was taken for

the ^{14}C measurements reported here. The core was cleaned, labeled, and immediately placed vertically in a chest freezer to eliminate microbial degradation and compaction due to de-watering. The second core (sixth placement) was for archive purposes and it was cleaned, labeled, and stored vertically in a walk-in refrigerator. Subcore material consisted of 10 cm i.d. thin-wall schedule 20 PVC pipe. The pipe was pre-cleaned by scrubbing with a soap solution, rinsing with 10% HCl, and rinsing with de-ionized water before insertion. The subcores were capped and sealed immediately.

The frozen subcores were split using a band saw at Woods Hole. Liquid nitrogen was used to keep the subcore frozen during splitting. Half of each subcore was wrapped immediately in plastic and returned to the freezer as an archive, while the working half of the subcore was first scraped with a clean knife to remove saw marks, described, sampled, and then returned to the freezer. Minor sediment disruption occurred due to ice crystal formation, but none of the fine-scale structure was obliterated and each of the horizons of ARTHUR *et al.* (in press) were readily identifiable. A sampling scheme for ^{14}C analysis established by M. Arthur and B. Hay to coordinate the radiocarbon and varve chronology work was used. Distinct horizons are easily seen and correlated (e.g. see Fig. 2 in HAY *et al.*, 1991). Each sample was limited to a 0.3–0.5 cm thickness. All equipment used for sampling was pre-cleaned with 10% HCl, rinsed with methanol and baked in a muffle furnace for 2 h @ 550°C. Each sample was placed in a 50°C oven to dry overnight. The samples were lightly crushed with a pre-cleaned mortar and pestle, placed in a vial, and stored in a desiccator. Each subcore sample interval chosen was prepared for AMS ^{14}C analysis at WHOI on the total carbonate or total organic carbon fractions by producing CO_2 either through hydrolysis (using approximately 50–200 mg of sediment) or closed-tube combustion (using approximately 20–100 mg of sediment) reactions then converted to graphite using methods outlined in GAGNON and JONES (1993). All radiocarbon analyses on material collected during the 1988 Black Sea Expedition were performed at the Tandem Accelerator Mass Spectrometry Facility located at the University of Arizona, Tucson, AZ. The pre-bomb mollusc shell was analysed at the National Ocean Sciences Accelerator Mass Spectrometry Facility located in Woods Hole, MA (JONES *et al.*, 1990).

Giant gravity cores

Nine of the 62 giant gravity cores collected during R.V. *Knorr* cruise 134, leg 1, were selected to provide Unit II sediments from as wide a water depth range as possible (Table 1 and Fig. 1). The cores selected were recovered from depths as shallow as 205 m and as deep as 2195 m. These cores were taken using the WHOI giant gravity coring system. This corer used up to 6 m long sections of 10 cm diameter schedule 40 PVC pipe. The corer was lowered at an average rate of 100 m min^{-1}, and the mean core length of recovered sediment exceeded 2.5 m. The cores were cut into 1.5 m long sections, immediately capped, sealed, labeled and stored vertically in a refrigerated van. The cores were split at WHOI and described by M. Arthur and B. Hay; who identified a series of distinct lithostratigraphic markers (defined as horizons A–L). Sampling for AMS ^{14}C analysis was done on the same day the cores were split and described. The samples were placed in plastic bags, sealed, and immediately frozen. Generation of CO_2 by hydrolysis or closed-tube combustion and subsequent conversion to graphite was performed as outlined in the previous section.

Seawater samples

Samples for determining the $\Delta^{14}C$ of dissolved inorganic carbon (DIC) were collected at sites BSK1 and BSK3 (Fig. 1 and Table 1) using a 12 sample rosette containing 30 l Niskin and/or Go-flo sample bottles. The rosette sampler was connected to a conductive winch cable permitting real-time CTD data acquisition. Prior to each deployment, the sample bottles were cleaned with a soap solution, rinsed with de-ionized water, 10% HCl, de-ionized water and finally with methanol.

Samples for $\Delta^{14}C$ analysis were always drawn first from the Niskin or Go-Flo bottles followed by sampling for a diverse set of physico-chemical measurements. To minimize the chance of contamination surgical rubber gloves were worn and fresh plastic was placed on all work surfaces at all times. Pre-cleaned (10% HCl, de-ionized water) silicon tubing with an in-line quartz fiber filter was connected to each bottle. A nitrogen gas cylinder was connected to the top of the Niskin or Go-Flo bottle and the gas pressure was used to force the seawater through the filter and into one of two sample bottles. A 1-l sample was collected in a pre-cleaned bottle (same procedure as above), poisoned with 3 mls of saturated mercuric chloride, and stored at ambient temperature. In addition, a 4-l sample was collected in a pre-cleaned bottle (soaked in Chromerge and baked for 2 h @ 550°C) and immediately placed in a chest freezer.

The 1-l samples were stripped of CO_2 gas at the Woods Hole Oceanographic AMS Graphite Preparation Lab, split into separate aliquots for $\delta^{13}C$ and $\Delta^{14}C$ analysis, and the CO_2 was converted to graphite (GAGNON and JONES, 1993). The 4-l samples were collected for making $\Delta^{14}C$ measurements of the DOC fraction and are presently stored frozen at the National Ocean Sciences AMS facility located at Woods Hole.

Sediment trap sampling

Sediment trap samples were collected to compare directly the $\Delta^{14}C$ of the water column DIC, the particulate organic matter (POM), and the most recently deposited sediments to better understand the residence times of Black Sea surface and deep waters. They were also made to examine the relationship between the $\Delta^{14}C$ of surface waters and the $\Delta^{14}C$ of the carbon fixed by plankton, as there is at present disagreement over these processes (e.g. MURRAY *et al.*, 1991). Three Mark 6 single-cup traps and three Mark 5-12 cup time-series traps were deployed during Leg 1 of R.V. *Knorr* cruise 134. Two of the Mark 5 traps were deployed at map code 4 (Fig. 1): one summer and one year-long deployment. All traps and trap cups used for $\Delta^{14}C$ analysis were washed with a soapy solution, rinsed with distilled water, sprayed with a 10% HCl solution, rinsed again with distilled water, and sprayed with methanol immediately prior to deployment. The Mark 5 summer sediment trap was deployed on 5 May, 1988 at a water depth of 1200 m and recovered on 14 July, 1988 by V. Asper aboard the R.V. *Knorr*. Four of the 12 trap cups (positions 2, 5, 9, 11) were allocated for AMS radiocarbon analysis. Although trap cups are filled routinely with a buffered 4% Formalin and 50% NaCl solution before deployment, Formalin was not included in the cups designated for ^{14}C analysis to reduce the possibility of organic carbon contamination. Clean plastic was spread on all work surfaces and rubber gloves were worn at all times to further reduce the chance of contamination. Upon recovery, the ^{14}C trap cups were removed first and immediately sealed and frozen.

In the laboratory, the trap cups were thawed at ambient room temperature, the

supernate measured for pH (nominally = 7.6) and discarded, and each sample was wet split into four equal sub-samples. Cups 2 and 11 contained the most material, and were chosen for $\Delta^{14}C$ analysis. Each sample split was filtered onto a pre-weighed and baked (600°C) quartz-fiber filter than dried in a 50°C oven. The dry filter and sample were reweighed and split for AMS graphitization, using half for $CaCO_3$ hydrolysis and half for organic carbon closed-tube combustion analysis as outlined in GAGNON and JONES (1993). The $CaCO_3$ hydrolysis of the sediment trap material proved unsuccessful due to a lack of biogenic carbonate. AMS ^{14}C analyses were performed at the Tandem Accelerator Mass Spectrometer Facility located at the University of Arizona, Tucson, Arizona.

RESULTS

Seawater samples and pre-bomb mollusc

Previous results from this basin have been reported in OSTLUND (1974) and OSTLUND and DYRSSEN (1986). OSTLUND and DRYSSEN (1986) questioned the radiocarbon data obtained during the 1965 *Odysseus* cruise to the Black Sea because of a suspected unreliable sampling system, and we do not discuss it further. The 1984 *Vityaz* cruise data reported in OSTLUND and DYRSSEN (1986) are included in Fig. 3. Surface values from our study clearly show the influence of bomb radiocarbon resulting from nuclear weapons testing in the atmosphere during the early-to-mid 1960s. Values for $\Delta^{14}C$ are as high as +120‰. The $\Delta^{14}C$ values decrease quite rapidly with water depth and are approximately −210‰ for the entire deep, anoxic part of the basin. The radiocarbon profiles from the two BSK stations agree closely for depths below approximately 200 m; however for depths shallower than 200 m, the BSK-1 values are approximately 50‰ lower than those of the BSK-3 site.

Our values are indistinguishable, within the analytical precision of the measurements, from those reported in OSTLUND and DRYSSEN (1986) for waters deeper than approximately 600 m; however our values are significantly lower for the shallower depths. Several explanations are possible, the most likely being the general trend for decreasing radiocarbon activity of surface waters of the world's ocean since the mid-1970s due to the mixing of radiocarbon-depleted waters from deeper depths with bomb ^{14}C enriched surface waters (e.g. TOGGWEILER *et al.*, 1989).

Unfortunately, due to the atmospheric nuclear weapons testing from the mid-1950s to the mid-1960s it is impossible today to measure directly the natural $\Delta^{14}C$ of the surface waters, a value essential for determining the reservoir-correction to be applied to the downcore radiocarbon analyses. We have determined this value, however, by measuring the radiocarbon activity of a shell of the bivalve mollusc *Mytilus galloprovincialis*. This shell was collected live from the Black Sea (map code 9) in 1931 by Dr. B. Tsvetkov and archived at the Moscow State University. This shell has a $\Delta^{14}C$ value of −55‰ or a conventional ^{14}C age of 460 ± 35 years. This directly measured value contrasts significantly with a pre-bomb surface value of −200‰ estimated by MURRAY *et al.* (1991) (Fig. 3).

Our assumed pre-bomb $\Delta^{14}C$ water column profile is presented in Fig. 3 as a dashed line. The profile of our measured values shows a shallow penetration of bomb ^{14}C into the Black Sea, which is consistent with the shallow permanent sharp halocline in this basin. Also, the rapid decrease in the $\Delta^{14}C$ values observed between the early 1980's data of Ostlund and Dyrssen and our late 1980s data is consistent with a rapid flushing of the surface water in

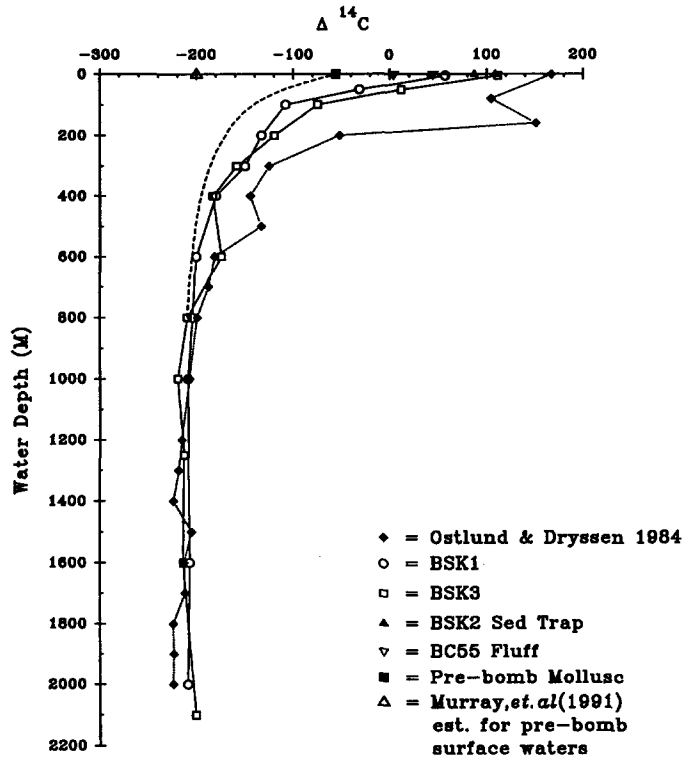

Fig. 3. Two water column profiles of DIC $\Delta^{14}C$ from the central Black Sea. Results are listed in Table 2 and follow the convention of STUIVER and POLACH (1977). Note internal consistency between the BSK1 and BSK3 profiles. Radiocarbon analyses of the Total Organic Carbon fraction from the "fluff" layer in BC-55, and particulate organic matter from BSK-2 sediment trap are consistent with the surface water $\Delta^{14}C$ values. Our data is in agreement with that of OSTLUND and DYRSSEN (1984) for those depths below approximately 600 m but is more depleted for those analyses shallower than 600 m. Mollusc-derived pre-bomb surface water value (this paper) and pre-bomb surface water estimate of MURRAY *et al.* (1991) are shown. Dashed line is our estimate of the pre-bomb water column profile.

this basin and rapid atmospheric equilibration of the surface waters. The 50‰ decrease in $\Delta^{14}C$ of surface water observed between 1984 (OSTLUND and DYRSSEN, 1986) and 1988 (BSK-3) is consistent with the 50‰ decrease in the $\Delta^{14}C$ of the atmosphere for this same period (BURCHULADZE *et al.*, 1989; POVINEC *et al.*, 1993). Furthermore, the surface water-atmosphere $\Delta^{14}C$ difference for these two times is −49‰ (1984) and −46‰ (1988), values that are consistent with the pre-bomb surface water value of −55‰.

Sediment trap samples

The $\Delta^{14}C$ values of the two sediment trap samples from a trap deployed at 1200 m depth at map code 4 were 110‰ (cup 2) and 87‰ (cup 11). The radiocarbon activity of the surface water samples were 60‰ for BSK-1 (to the west) and 115‰ for BSK-3 (to the east). These samples were processed as described in the methods section, and the results show that the radiocarbon activity of particulate organic matter is intermediate between the lower

Table 2. Results of AMS Radiocarbon Analyses on Seawater, Sediment Trap, pre-bomb mollusc and "Fluff" sediments. Seawater sample analyses were made on the Dissolved Inorganic Carbon (DIC) fraction. The sediment trap and Fluff sample analyses were made on the Total Organic Carbon (TOC) fraction. Results are reported according to the convention of STUIVER and POLACH (1977). Values for $\delta^{13}C$ were measured from CO_2 gas evolved from the sample using a VG Prism Stable Isotope Mass Spectrometer, except those annotated with (*) CO_2 from recombusting graphite, (†) interpolated from surrounding or (‡) assumed measured values. Accession numbers are for the sample preparation (WHG #s) and the AMS radiocarbon analyses (AA or OS #s)

Sample ID	Sample type	Water depth (m)	Total CO_2 (mM kg^{-1})	$\delta^{13}C$ wrt PDB	$\Delta^{14}C$	Arizona accession no.	WHOI accession no.
KNR134-08 BSK-1	DIC	5.0	3.256	−1.98	57.3	AA4015	WHG-321
KNR134-08 BSK-1	DIC	50.0	3.266	−2.02	−31.5	AA4173	WHG-322
KNR134-08 BSK-1	DIC	100.0	3.376	−2.83	−108.1	AA4183	WHG-354
KNR134-08 BSK-1	DIC	200.0	3.495	−3.50	−132.9	AA4016	WHG-339
KNR134-08 BSK-1	DIC	300.0	3.624	−4.19	−149.8	AA4186	WHG-369
KNR134-08 BSK-1	DIC	400.0	3.837	−5.25	−180.2	AA4181	WHG-346
KNR134-08 BSK-1	DIC	600.0	3.960	−5.76	−200.5	AA4179	WHG-324
KNR134-08 BSK-1	DIC	800.0	3.944	−5.99	−204.1	AA4184	WHG-363
KNR134-08 BSK-1	DIC	1000.0	3.895	−6.24	−208.8	AA4180	WHG-335
KNR134-08 BSK-1	DIC	1600.0	4.307	−6.56	−207.3	AA3812	WHG-319
KNR134-08 BSK-1	DIC	2000.0	4.083	−6.60	−209.2	AA4182	WHG-349
KNR134-08 BSK-3	DIC	5.0	3.244	−1.21	111.6	AA4195	WHG-375
KNR134-08 BSK-3	DIC	50.0	3.293	−2.24	11.9	AA4196	WHG-390
KNR134-08 BSK-3	DIC	100.0	3.371	−2.92	−74.9	AA4197	WHG-381
KNR134-08 BSK-3	DIC	200.0	3.418	−4.78*	−119.6	AA4200	WHG-403
KNR134-08 BSK-3	DIC	300.0	3.607	−5.53*	−158.6	AA4202	WHG-387
KNR134-08 BSK-3	DIC	400.0	3.829	−5.22	−183.2	AA4203	WHG-378
KNR134-08 BSK-3	DIC	600.0	3.793	−5.04	−174.5	AA4204	WHG-373
KNR134-08 BSK-3	DIC	800.0	4.053	−5.74†	−209.6	AA4205	WHG-399
KNR134-08 BSK-3	DIC	1000.0	4.123	−6.45	−219.5	AA4206	WHG-376
KNR134-08 BSK-3	DIC	1250.0	4.145	−6.73	−213.0	AA4207	WHG-388
KNR134-08 BSK-3	DIC	1600.0	4.144	−6.78†	−214.0	AA4208	WHG-379
KNR134-08 BSK-3	DIC	2100.0	3.992	−6.82	−200.8	AA4210	WHG-372
KNR134-08 BSK-2 Sed Trap	TOC	Cup no. 11	—	−25.00‡	87.1	AA5365	WHG-716
KNR134-08 BSK-2 Sed Trap	TOC	Cup no. 2	—	−25.00‡	109.0	AA5366	WHG-717
KNR134-08 BC55 Fluff	TOC	2164.0	—	−25.00‡	44.6	AA4728	WHG-505
KNR134-08 BC55 Fluff	TCC	2164.0	—	0.00‡	3.9	AA4642	WHG-504
Mytilus galloprovincialis	TCC	<5.0	—	0.72	−54.8	—	OS-718

activity to the west and the higher activity to the east. These data suggest that the organic matter being produced in the Black Sea is in approximate equilibrium with the DIC of the water in which it was produced (Table 2 and Fig. 3).

Sediment samples

All three box cores studied (Tables 1 and 3 and Fig. 1) contained the entire Unit I. HAY *et al.* (1991) have shown that within Unit I distinctive packets of laminae can be correlated

across the entire basin and five such intervals were chosen for analysis based upon visual inspection of each core. Box-core BC55 (Fig. 1, map code 7) was chosen as our reference core because it is one of the few containing both a well-preserved "fluff" layer and exhibiting no evidence of turbidites within Unit I. The carbonate and organic carbon fractions of the "fluff" layer contain bomb ^{14}C (Table 3), and the presence of shorter half-life isotopes such as ^{210}Pb (CRUSIUS and ANDERSON, 1991) and other radionuclides in the "fluff" layer of other cores (MOORE and O'NEILL, 1991) demonstrate that the most recently deposited sediments can be recovered successfully by the box-coring methods used. The five Unit I horizons sampled (Fig. 4) show a consistent pattern of increasing radiocarbon age with depth in all three cores, and core-to-core comparisons show internal consistencies, in both the organic carbon and carbonate fractions. The non-corrected radiocarbon age of Horizon-E is 2320 years BP based upon the carbonate fraction and 2390 years BP based upon the organic carbon fraction. The non-corrected radiocarbon age of Horizon-G is 3330 years BP for the carbonate fraction and 3150 years BP for the organic carbon fraction. In the discussion section we refine these ages by taking into account the reservoir correction, the effects of detrital organic carbon and carbonate, and converting the radiocarbon ages to calendar ages to derive a best estimate of 1635 ± 60 years BP for Horizon-E and 2720 ± 160 years BP for Horizon-G.

The giant gravity cores (GGCs) were selected based upon location, water depth and visual inspection of each core. Our strategy for sampling the GGC's was to analyse the same lithostratigraphic horizon in each of the cores for both carbonate and organic carbon radiocarbon. The lithostratigraphic determinations were made independently by M. Arthur and B. Hay (ARTHUR *et al.*, in press). Unlike Unit I, the results from Unit II exhibit carbonate-fraction radiocarbon ages that are substantially older than the organic carbon fraction from the same sample (Table 3, Fig. 5). For the organic carbon radiocarbon analyses, ages increase systematically with depth in core (Fig. 5) and the ages obtained on each of the lithostratigraphic horizons are approximately the same irrespective of water depth (Fig. 6). In contrast, the ages obtained on the carbonate fraction do not consistently increase the increasing depth in core (Fig. 5) and there is much less core-to-core consistency in the ages for each of the lithostratigraphic horizons (Fig. 6). The only exception to this pattern is Horizon-K, a chemically precipitated aragonite (Ross and DEGENS, 1974). Both the organic carbon and the carbonate radiocarbon ages for this horizon agree closely for those cores recovered from shallow water depths (Fig. 6).

DISCUSSION

Age of Unit I sediments

The initial estimate for the age of Horizon-E was made on a single conventional radiocarbon analysis of total organic carbon from sediments recovered during the 1969 R.V. *Atlantis II* expedition (Ross and DEGENS, 1974). The 3090 years BP age of this horizon was later questioned by DEGENS *et al.* (1980) who counted the laminations preserved in Black Sea sediments, and, by assuming they were annual, concluded that the age of the horizon was only 998 years BP. Several hypotheses can be put forth to reconcile the radiocarbon result of Ross and DEGENS (1974) and the varve chronology of DEGENS *et al.* (1980) and the data presented here can be used to test each of these hypotheses to derive an optimal chronology for these sediments.

Table 3. *Results of AMS Radiocarbon analyses on Unit I sediments from KNR*134-08 *box cores. Locations of cores are given in Table* 1 *and shown in Fig.* 1. *TOC = Total Organic Carbon fraction. TCC = Total Carbonate fraction. Lithostratigraphic horizons are those identified by* ARTHUR et al. (in press) and shown in *Fig.* 2. *Radiocarbon ages are corrected for reservoir effects* (60 years *for TOC*, 460 years *for TCC*), *detrital carbon input* (580 years *for TOC*, 260 years *for TCC*), *and converted to calendar age using* STUIVER *and* BRAZIUNAS (1993). *The* 1-σ *error for the calibrated ages were determined by adding in quadrature all sources of error including the error of the conventional radiocarbon measurement, and uncertainties in the pre-bomb reservoir age, detrital correction and the calibration dataset. An asterisk* (*) *after the core depth indicates an analysis that was not used in our calculations. No age listed under the reservoir correction, detrital correction or calendar calibration columns indicates an age that is corrected to a modern* (i.e. 0 years BP) *value*

Core	Depth interval (cm)	Sample type	Lith. horizon	Conventional age (years BP)	Reservoir corrected age (years BP)	Reservoir and detrital corrected age (years BP)	Calibrated age (cal BP)	Arizona accession no.	WHOI accession no.
KNR134-08 BC17	0.7–1.4	TOC	B	825 ± 45	765	185	175 ± 140	AA4721	WHG-516
KNR134-08 BC17	11.0–11.4	TOC	C	1180 ± 50	1120	540	550 ± 95	AA4967	WHG-550
KNR134-08 BC17	20.6–21.1	TOC	D	1910 ± 60	1850	1270	1200 ± 130	AA4732	WHG-517
KNR134-08 BC17	27.3–27.8	TOC	E	2430 ± 50	2370	1790	1760 ± 150	AA4968	WHG-551
KNR134-08 BC17	33.8–34.3	TOC	G	3640 ± 70	3580	3000	3240 ± 170	AA4733	WHG-518
KNR134-08 BC17	0.7–1.4	TCC	B	1000 ± 100	540	280	300 ± 100	AA4637	WHG-498
KNR134-08 BC17	11.0–11.4	TCC	C	960 ± 65	500	240	270 ± 140	AA4638	WHG-499
KNR134-08 BC17	20.6–21.1	TCC	D	1790 ± 65	1330	1070	1030 ± 140	AA4639	WHG-500
KNR134-08 BC17	27.3–27.8	TCC	E	2170 ± 90	1710	1450	1400 ± 150	AA4640	WHG-501
KNR134-08 BC17	33.8–34.3	TCC	G	3350 ± 70	2890	2630	2830 ± 170	AA4641	WHG-503

KNR134-08 BC25	0.3–0.8	TOC	B	390 ± 50	330	—	—	AA5028	WHG-610
KNR134-08 BC25	11.5–12.0	TOC	C	980 ± 50	920	340	380 ± 100	AA5018	WHG-615
KNR134-08 BC25	22.5–23.0	TOC	D	1890 ± 50	1830	1250	1180 ± 120	AA5021	WHG-612
KNR134-08 BC25	41.0–41.5	TOC	E	2310 ± 55	2250	1670	1640 ± 150	AA5025	WHG-616
KNR134-08 BC25	48.8–49.3	TOC	G	3000 ± 55	2940	2360	2500 ± 160	AA5019	WHG-617
KNR134-08 BC25	0.3–0.8	TCC	B	1270 ± 50	810	550	550 ± 100	AA5012	WHG-602
KNR134-08 BC25	11.5–12.0	TCC	C	1370 ± 50	910	650	610 ± 100	AA5013	WHG-603
KNR134-08 BC25	22.5–23.0	TCC	D	1990 ± 50	1530	1270	1200 ± 130	AA5014	WHG-606
KNR134-08 BC25	41.0–41.5	TCC	E	2490 ± 50	2030	1770	1740 ± 150	AA5015	WHG-607
KNR134-08 BC25	48.8–49.3	TCC	G	3540 ± 50	3080	2820	3020 ± 160	AA5016	WHG-608
KNR134-08 BC55	0.0–0.7	TOC	A	−390 ± 55	—	—	—	AA4728	WHG-505
KNR134-08 BC55	2.9–3.5	TOC	B	840 ± 100	780	200	210 ± 150	AA4215	WHG-411
KNR134-08 BC55	14.0–14.6	TOC	C	1120 ± 100	1060	480	480 ± 130	AA4214	WHG-406
KNR134-08 BC55	24.2–24.6	TOC	D	1990 ± 70	1930	1350	1290 ± 140	AA4220	WHG-407
KNR134-08 BC55	32.2–32.6	TOC	E	2420 ± 60	2360	1780	1750 ± 150	AA4222	WHG-409
KNR134-08 BC55	39.5–39.7*	TOC	G	845 ± 55	785	205	210 ± 130	AA4223	WHG-410
KNR134-08 BC55	39.2–39.6	TOC	G	2810 ± 65	2750	2170	2200 ± 170	AA4729	WHG-506
KNR134-08 BC55	0.0–0.7	TCC	A	−70 ± 65	—	—	—	AA4642	WHG-504
KNR134-08 BC55	2.9–3.5	TCC	B	860 ± 45	400	140	160 ± 110	AA4006	WHG-386
KNR134-08 BC55	14.0–14.6	TCC	C	1000 ± 100	540	280	320 ± 150	AA4213	WHG-385
KNR134-08 BC55	24.2–24.6	TCC	D	1830 ± 100	1370	1110	1060 ± 150	AA4212	WHG-384
KNR134-08 BC55	32.2–32.6	TCC	E	2290 ± 100	1830	1570	1520 ± 170	AA4211	WHG-382
KNR134-08 BC55	39.5–39.7	TCC	G	3100 ± 100	2640	2380	2520 ± 180	AA4217	WHG-380

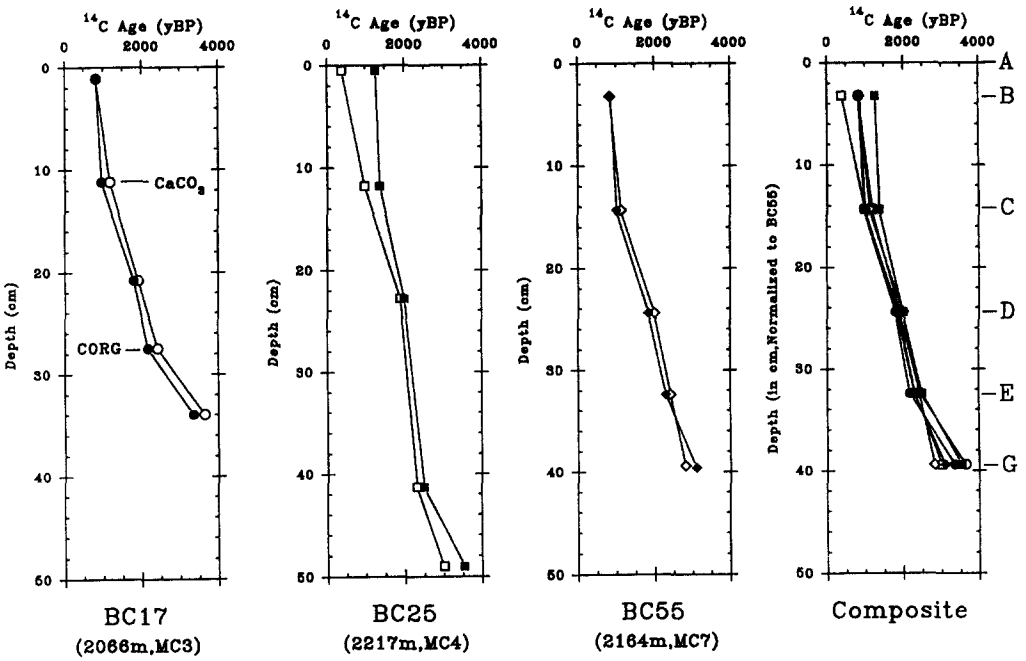

Fig. 4. Conventional radiocarbon age–depth plots for the Total Organic Carbon (open symbols) and Total Carbonate (closed symbols) fractions from Unit I sediments in three boxcores. The same lithostratigraphic horizons were sampled in each of the three cores. The composite plot uses a common (BC55) depth scale to facilitate core-to-core comparison of the ages. The corresponding lithostratigraphic horizons of ARTHUR *et al.* (in press) are shown on the composite age–depth plot. Ages from each core are listed in Table 3. Core locations are given in Table 1 and the corresponding map code is shown in Fig. 1.

One explanation to reconcile the radiocarbon results of Ross and DEGENS (1974) and the varve chronology of DEGENS *et al.* (1980) is that the upper 2100 years of sediment were lost during the coring process. The sediments analysed for those earlier studies were recovered by piston coring which commonly does not recover the true core top. In addition, recovery of true core top material in Black Sea sediments is difficult because of the high water content of the upper several centimeters. Therefore, if Horizon-E is in fact 3100 years BP and 2100 years of the sediment record are missing due to the coring process, the radiocarbon ages should be 2100 years old at the recovered core top, 3100 years BP at Horizon-E and there would be 1000 laminations between the coretop and Horizon-E.

A second explanation to reconcile these different chronologies was put forth by DEGENS *et al.* (1980). These authors argued that the radiocarbon chronology was too old by approximately 2000 years due to the input of detrital carbon. Detrital input of carbonate has been shown by Ross and DEGENS (1974) to influence clearly the radiocarbon age of Unit II sediments, and TRIMONIS (1974) identified a number of potential detrital carbonate sources around the Black Sea. Although coretop sediments were not analysed by Ross and DEGENS (1974), CALVERT (1987) did look at coretop sediments from these cores and found the ages to be approximately 2000 years BP. From these data alone, the discrepancy between Ross and DEGENS (1974) and DEGENS *et al.* (1980) cannot be resolved.

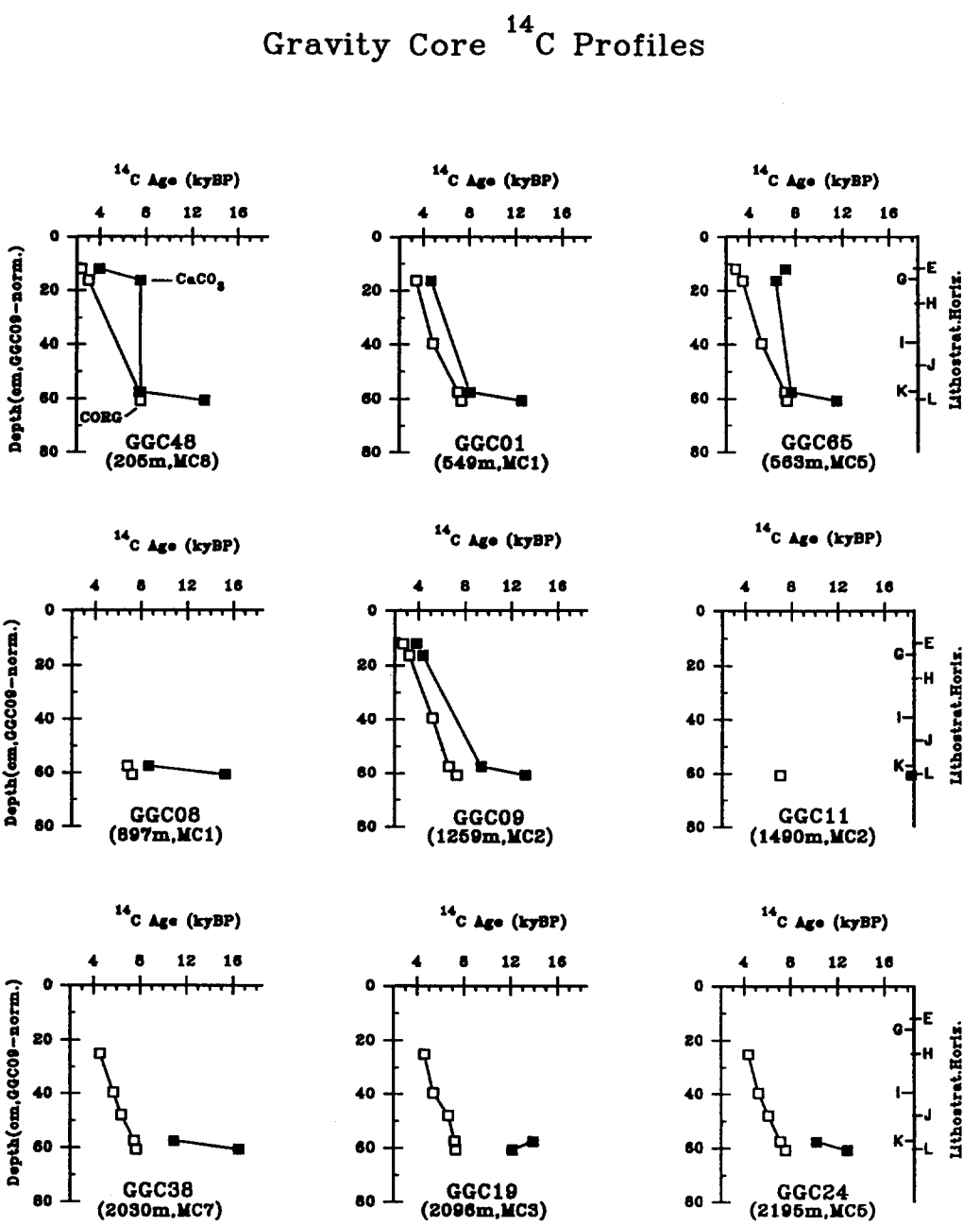

Fig. 5. Conventional radiocarbon age–depth plots for all Giant Gravity Cores studied. Depths have been normalized to those of core GGC09. True age–depth plots for each core can be constructed from data listed in Table 4. Corresponding lithostratigraphic horizons are shown on right. Open symbols = Total Organic Carbon fraction analyses. Solid symbols = Total Carbonate fraction analyses. Ages are consistently younger for the organic carbon analyses than for the total carbonate analyses. The corresponding map code (MC) for each core is given.

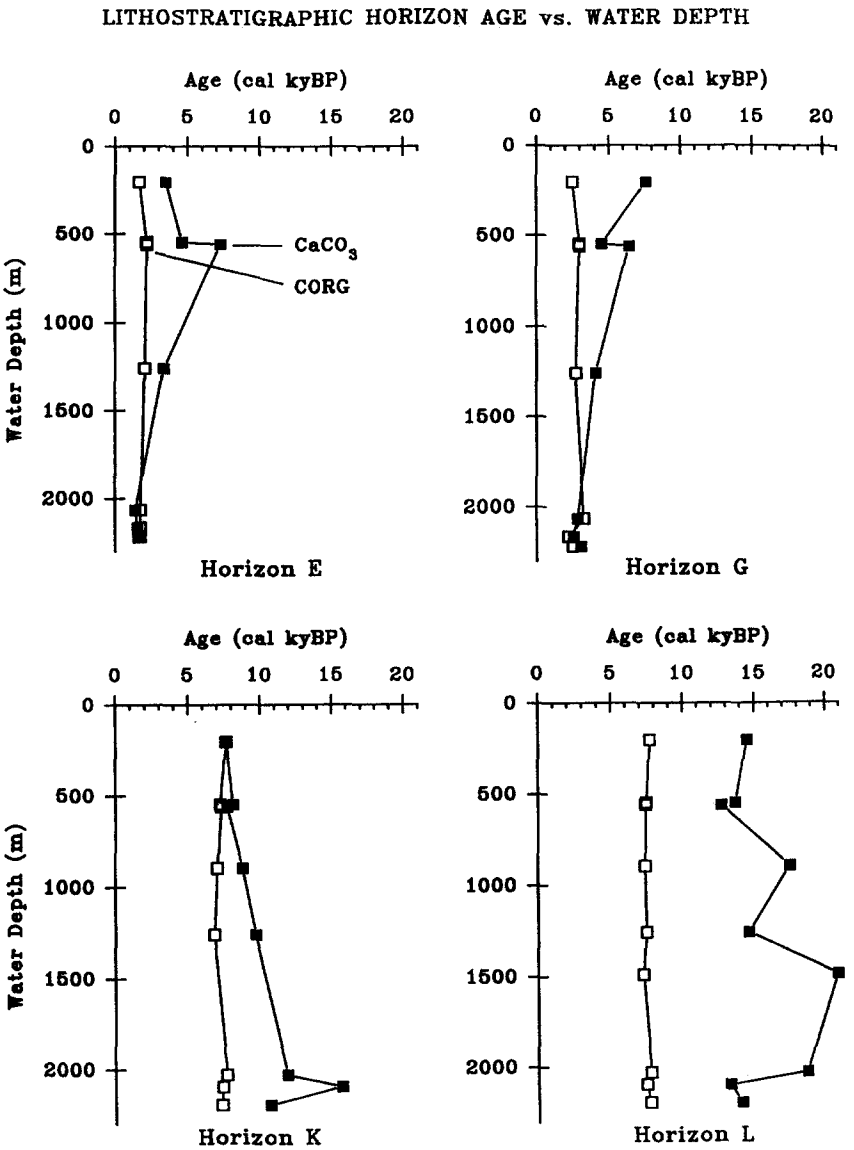

Fig. 6. Calibrated radiocarbon ages (calendar years) of four lithostratigraphic horizons vs water depth. There is no significant change in the age of the horizon with water depth for the Total Organic Carbon fraction (open symbols), but a great deal of scatter is observed for the Total Carbonate analyses (closed symbols).

These two hypotheses can be tested, however, by analysing sediments from cores with well-documented coretop recovery. The cores obtained during the 1988 expedition were carefully collected so as to prevent the disturbance and (or) loss of coretop material by the coring process. All of the radiocarbon ages obtained on both the carbonate and organic carbon fractions from sediments in the upper 25 cm are younger than 2000 years BP (Table

2), and are consistent with AMS radiocarbon analyses of the organic carbon fraction from box cores collected by CALVERT *et al.* (1991). Both datasets show that the sediment–water interface was not recovered in those cores collected during the 1969 and 1974 cruises, and refutes the contention of DEGENS *et al.* (1980) that the contribution of detrital carbon (or carbonate) to these sediments increases the radiocarbon ages by 2000 years. In addition, AMS radiocarbon dating of the carbonate and organic carbon fractions of the surfacemost "fluff" layer in box core BC55 shows that this material was deposited sometime after the mid 1960s, as it contains bomb ^{14}C. Lead-210 (CRUSIUS and ANDERSON, 1991) and radionuclide (MOORE and O'NEILL, 1991) analyses from other box cores collected during the 1988 expedition independently show that this "fluff" layer was deposited in the last several years, confirming that true core tops have been recovered. Extrapolation of the sedimentation rates calculated by the ^{210}Pb method for the upper few centimeters (CRUSIUS and ANDERSON, 1992) are consistent with the approximate 20 cm 10^{-3} year sedimentation rates obtained from our radiocarbon analyses for Unit I sediments between Horizons B and E.

Knowing that the most recently deposited material has been collected, it should be a simple matter to resolve the varve vs radiocarbon chronology controversy. Before doing this, however, we need to apply three corrections to the radiocarbon ages reported in Table 2. First, the $\delta^{13}C$ of the carbonate fraction from Unit I sediments has a value near 0 per mil (PDB) (Arthur, personal communication) and the $\delta^{13}C$ of the organic matter has a value near -24 per mil (PDB) (CALVERT and FONTUGNE, 1987). The convention for reporting radiocarbon ages is to normalize to a $\delta^{13}C$ value of -25 per mil (PDB) resulting in an apparent 400 year age for modern marine carbonates precipitated in pre-bomb waters. Although this reservoir correction for marine carbonates averages 400 years, it has been shown to vary from approximately 200 years to over 800 years depending on geographic location and oceanographic setting (e.g. STUIVER and BRAZIUNAS, 1993). MURRAY (1991) has speculated that the pre-bomb apparent age of the surface waters of the Black Sea was 1430 years. This value is in error for two reasons; first, all radiocarbon ages of near-surface sediment from box cores are younger than this and second, the age of a mollusc shell collected live in 1931 was dated to 460 ± 35 years BP (Table 2). The 460 year age (-55‰) is a direct estimate of the $\Delta^{14}C$ of the pre-bomb surface waters of the Black Sea. We thus apply a 460 and 60 year (460–400) reservoir correction to the carbonate and organic carbon fractions of the Black Sea sediment analysed.

Second, we must obtain an estimate of the detrital carbon (carbonate) correction for Unit I sediments. Unfortunately, the best preserved cores contain bomb-^{14}C in the core top sediments (Horizon A) and we cannot obtain these corrections by direct measurement of the core top ^{14}C ages. Rather we determine this value by using the 15 pairs of pre-bomb reservoir-corrected organic carbon-carbonate radiocarbon analyses listed in Table 3. These data show the total organic carbon fraction to be, on average, 320 years older than the total carbonate fraction for Unit I sediments. We adjust the organic carbon ages by this amount. The reservoir-corrected and organic carbon adjusted radiocarbon ages from each of the three cores analysed are determined for each horizon (six measurements per horizon). We then extrapolate using the depths of the Unit I horizons in BC-55, the core with the best preserved sediment–water interface, to determine the Horizon A (i.e. 0 cm) age of 260 years. We consider 260 years and 580 years (i.e. 260 + 320) to be the maximum detrital correction that should be applied to Unit I carbonates and organic carbon sediments, respectively.

A third correction is needed to convert from a radiocarbon age to a calendar age since varves are by definition a calendar chronology. Radiocarbon chronology is not an exact calendar chronology because of fluctuations in the rate of cosmogenic production of ^{14}C. A calibration dataset has been assembled and modeled in order to convert marine radiocarbon ages to calendar years (STUIVER and BRAZIUNAS, 1993). We calculate the calendar ages of the Black Sea sediments by using ΔR (i.e. local surface water value of 460 years for carbonate or 60 years for organic carbon minus the global average surface water value of 400 years) values of +60 and −340 years for carbonate and organic carbon samples, respectively, and the calibration curves presented in STUIVER and BRAZIUNAS (1993) (Table 3). Applying all three corrections leads us to estimate Horizon-E as 1635 ± 60 years BP and Horizon-G as 2720 ± 160 years BP.

ARTHUR *et al.* (in press) report 1320 light-dark couplets preserved in box-core recovered sediments above Horizon-E. This is more than measured by DEGENS *et al.* (1980), but still 20% less than the radiocarbon analyses would suggest. Examination of the number of couplets in Unit I down to Horizon-G (ARTHUR *et al.*, in press) shows that 1700 couplets are preserved in Unit I sediments from cores exhibiting excellent coretop preservation. This estimate is 35% less than our age estimate for Horizon-G. Given that our estimate already incorporates reservoir, detrital, and radiocarbon-to-calendar corrections, we suggest that the varve counts underestimate the age of Unit I. This underestimate would occur if in some years the couplets formed are extremely thin and difficult to resolve (ARTHUR *et al.*, in press; CRUSIUS and ANDERSON, 1992).

The possibility also exists that couplets are not formed every year, for example, if a coccolith or diatom bloom did not occur in a particular year. Results of sediment trapping have shown that in only four of five years studied would a visually identifiable couplet been preserved in the Black Sea sediments (Hay, personal communication), resulting in a 20% undercount, which is consistent with the radiocarbon/varve difference just discussed. Furthermore, Honjo (personal communication) has shown that the 20% undercount extends to an approximate 10 year long record of sediment trap results from the Black Sea. Considering all factors we feel that our standard error age estimate of 2720 ± 160 years, determined from the six radiocarbon measurements of Horizon-G listed in Table 3, for the first invasion of *E. huxleyi* and 1635 ± 160 years BP, determined from the six radiocarbon measurements of Horizon-E listed in Table 3, for the beginning of the last invasion of *E. huxleyi* are the best presently available.

Age of Unit II sediments

Unit II sediments are finely-laminated, carbonate-poor sapropelic muds deposited under anoxic conditions. Ross and DEGENS (1974) determined the age of Horizon-L to be 7090 ± 180 years from a single organic carbon radiocarbon measurement, and DEGENS *et al.* (1980) determined the age of this horizon to be 5080 years based upon varve counts from a single core. This 2000 year age difference is about the same as that reported by DEGENS *et al.* (1980) for Horizon-E. Correct determination of the age of this unit boundary has importance in understanding the timing of this basin's transition from a lacustrine to a marine system and its relation to sea-level rise. It is also needed to constrain the several hypotheses proposed for the spatial and temporal evolution of anoxia within the basin (e.g. DEUSER, 1974; GLENN and ARTHUR, 1985; CALVERT, 1987). We have selected nine of the giant gravity cores collected during leg 1 of the 1988 Black Sea expedition, from the

depth range 205 m to 2195 m (Tables 1 and 4). Unit II sediments are divided into six lithostratigraphic horizons ranging from G to L (ARTHUR *et al.*, in press). The conventional radiocarbon age and standard error of Horizon-L as determined by nine organic carbon radiocarbon analyses is 7350 ± 80 years BP (Table 4), which is in close agreement with the single radiocarbon analyses for this Unit boundary reported in Ross and DEGENS (1974) and CALVERT (1987).

To make core-to-core comparisons of the radiocarbon ages of each of the different lithostratigraphic horizons, we have normalized all of the age–depth plots to the horizon depths found in core GGC09 (map code 2 and Fig. 5). Plots of age vs true depth for each core can be constructed from the data in Table 4. Core GGC09 (map code 2) was chosen as the reference core because it is from the middle of the range of water depths analysed and it has the median sedimentation rate of all of the GGCs analysed. Even after applying the 460 year reservoir correction to the carbonate fraction and 60 years to the organic carbonate fraction radiocarbon ages as discussed in the previous section, the carbonate ages are significantly older than the organic carbon radiocarbon ages for nearly every Unit II horizon dated. This result contrasts with that in Unit I sediments, in which the reservoir-corrected ^{14}C ages for the carbonate fraction are on average 320 years younger than the corresponding organic carbon ages (Table 3 and Fig. 4). The main reason for this difference is the majority of the carbonates in Unit II sediments are reworked (TRIMONIS, 1974) whereas, only approximately 5% of the Unit I carbonates are reworked (HAY *et al.*, 1991).

The age–depth plots for the organic carbon ^{14}C analyses of Unit II sediments show no age reversals and suggest a nearly uniform sedimentation rate throughout the unit within each of the cores, although there are regional core-to-core differences in the sedimentation rates (Table 4). Plotting the organic carbon and carbonate radiocarbon ages by water depth for each of the lithostratigraphic horizons shows uniform ages of the organic carbon fraction for each horizon irrespective of water depth (Fig. 6). This pattern suggests that the major source of organic carbon to the sediments of the Black Sea during Unit II time is biologically produced *in situ* in the surface waters and that there is little reworking and lateral advection of detrital organic carbon off the shelves and into the basin.

For Unit I sediments we could estimate the reservoir and detrital carbon and carbonate corrections directly by studying a live-collected pre-bomb mollusc shell and the coretop sediments. For Unit I we cannot determine the reservoir correction directly but Horizon-K can be used to estimate the difference in the carbonate and organic carbon corrections. Horizon-K, an inorganically-precipitated aragonite, offers an excellent check on the detrital contribution to the carbonate and organic carbon radiocarbon ages. The aragonite should have a radiocarbon value in equilibrium with the seawater in which it was precipitated. We assume the equilibrium value is 460 years as measured from the live-collected pre-bomb mollusc discussed earlier. Unlike the organic carbon ages for Horizon-K, there is a trend toward increasing age with increasing water depth for the carbonate fraction. This pattern is most easily explained by the lateral advection of a detrital carbonate source off the shelf and slope and transport into the basin along isopycnals (e.g. HONJO *et al.*, 1982, JONES, 1984, HAY, 1987). There is excellent agreement between the two fractions in the shallowest cores where the cumulative effect of laterally advected detrital carbonate contribution to the sediments is at a minimum and the carbonate radiocarbon age is most closely recording the equilibrium value.

The inorganically-precipitated aragonite conventional ^{14}C age for the shallowest core,

Table 4. Results of AMS Radiocarbon analyses on Unit I and II sediments from KNR134-08 Giant Gravity Cores. Locations of cores are given in Table 1 and shown in Fig. 1. Description and treatment of samples is the same as for Table 3

Core	Depth interval (cm)	Sample type	Lith. horizon	Conventional age (years BP)	Reservoir corrected age (years BP)	Reservoir and detrital corrected age (years BP)	Calibrated age (cal BP)	Arizona accession no.	WHOI accession no.
KNR134-08 GGC01	47.5–48.0	TOC	E	2760 ± 60	2700	2120	2180 ± 190	AA5862	WHG-832
KNR134-08 GGC01	64.8–65.3	TOC	G	3410 ± 60	3350	2770	2950 ± 160	AA5863	WHG-845
KNR134-08 GGC01	115.3–115.8	TOC	I	4850 ± 80	4790	4210	4790 ± 190	AA5864	WHG-839
KNR134-08 GGC01	154.8–155.3	TOC	K	7010 ± 100	6950	6370	7250 ± 150	AA5442	WHG-683
KNR134-08 GGC01	158.8–159.3	TOC	L	7300 ± 80	7240	6660	7500 ± 120	AA5460	WHG-684
KNR134-08 GGC01	47.5–48.0	TCC	E	4770 ± 80	4310	4050	4620 ± 170	AA5858	WHG-820
KNR134-08 GGC01	64.8–65.3	TCC	G	4680 ± 50	4220	3960	4490 ± 180	AA5859	WHG-821
KNR134-08 GGC01	154.8–155.3	TCC	K	8040 ± 100	7580	7320	8130 ± 150	AA5438	WHG-672
KNR134-08 GGC01	158.8–159.3	TCC	L	12500 ± 130	12040	11780	13700 ± 220	AA5464	WHG-673
KNR134-08 GGC08	64.0–64.5	TOC	K	6840 ± 110	6780	6200	7060 ± 160	AA5439	WHG-674
KNR134-08 GGC08	68.3–68.7	TOC	L	7210 ± 70	7150	6570	7420 ± 120	AA5456	WHG-675
KNR134-08 GGC08	64.0–64.5	TCC	K	8640 ± 130	8180	7920	8790 ± 240	AA5434	WHG-665
KNR134-08 GGC08	68.3–68.7	TCC	L	15300 ± 140	14840	14580	17500 ± 210	AA5453	WHG-666
KNR134-08 GGC09	11.8–12.3	TOC	E	2660 ± 60	2600	2020	2040 ± 160	AA5865	WHG-841
KNR134-08 GGC09	16.0–16.5	TOC	G	3210 ± 50	3150	2570	2710 ± 160	AA5872	WHF-842
KNR134-08 GGC09	39.3–39.8	TOC	I	5220 ± 60	5160	4580	5290 ± 180	AA5873	WHG-843
KNR134-08 GGC09	57.3–57.8	TOC	K	6610 ± 100	6550	5970	6830 ± 170	AA5440	WHG-676
KNR134-08 GGC09	60.5–61.0	TOC	L	7290 ± 120	7230	6650	7490 ± 150	AA5465	WHG-677
KNR134-08 GGC09	11.8–12.3	TCC	E	3800 ± 50	3340	3080	3340 ± 160	AA5860	WHG-822
KNR134-08 GGC09	16.0–16.5	TCC	G	4360 ± 50	3900	3640	4050 ± 170	AA5867	WHG-824
KNR134-08 GGC09	57.3–57.8	TCC	K	9390 ± 130	8930	8670	9700 ± 180	AA5435	WHG-667
KNR134-08 GGC09	60.5–61.0	TCC	L	13200 ± 80	12740	12480	14600 ± 220	AA5868	WHG-825
KNR134-08 GGC11	4.0–4.5	TOC	L	7050 ± 80	6990	6410	7280 ± 120	AA5466	WHG-681
KNR134-08 GGC11	4.0–4.5	TCC	L	18300 ± 230	17780	17520	20900 ± 365	AA5463	WHG-663
KNR134-08 GGC19	18.5–19.0	TOC	H	4620 ± 70	4560	3980	4530 ± 190	AA4734	WHG-519
KNR134-08 GGC19	24.2–24.7	TOC	I	5380 ± 70	5320	4740	5470 ± 150	AA4735	WHG-520
KNR134-08 GGC19	31.0–31.5	TOC	J	6640 ± 60	6580	6000	6860 ± 150	AA4741	WHG-523

KNR134-08 GGC19	37.3–37.8	TOC	K	7200 ± 80	7140	6560	7410 ± 120	AA5459	WHG-682
KNR134-08 GGC19	39.0–39.5	TOC	L	7290 ± 90	7230	6650	7490 ± 130	AA4730	WHG-510
KNR134-08 GGC19	37.3–37.8	TCC	K	13900 ± 100	13440	13180	15700 ± 240	AA5457	WHG-679
KNR134-08 GGC19	39.0–39.5	TCC	L	12100 ± 120	11640	11380	13300 ± 150	AA4643	WHG-508
KNR134-08 GGC24	49.8–50.2	TOC	H	4370 ± 60	4310	3730	4170 ± 180	AA5887	WHG-846
KNR134-08 GGC24	61.5–62.0	TOC	I	5240 ± 60	5180	4600	5330 ± 180	AA5888	WHG-847
KNR134-08 GGC24	70.2–70.7	TOC	J	6100 ± 60	6040	5460	6270 ± 140	AA5945	WHG-889
KNR134-08 GGC24	78.8–79.3	TOC	K	7130 ± 60	7070	6490	7350 ± 120	AA5889	WHG-848
KNR134-08 GGC24	83.2–83.7	TOC	L	7560 ± 70	7500	6920	7740 ± 130	AA5890	WHG-849
KNR134-08 GGC24	78.8–79.3	TCC	K	10200 ± 70	9740	9480	10700 ± 190	AA5892	WHG-826
KNR134-08 GGC24	83.2–83.7	TCC	L	12820 ± 80	12360	12100	14100 ± 190	AA5893	WHG-827
KNR134-08 GGC38	17.3–17.8	TOC	H	4560 ± 60	4500	3920	4430 ± 190	AA5454	WHG-668
KNR134-08 GGC38	24.0–24.5	TOC	I	5690 ± 70	5630	5050	5810 ± 150	AA5455	WHG-669
KNR134-08 GGC38	31.1–31.6	TOC	J	6390 ± 70	6330	5750	6575 ± 150	AA5002	WHG-622
KNR134-08 GGC38	36.5–37.0	TOC	K	7500 ± 110	7440	6860	7690 ± 150	AA5436	WHG-670
KNR134-08 GGC38	38.2–38.7	TOC	L	7640 ± 80	7580	7000	7800 ± 130	AA4731	WHG-512
KNR134-08 GGC38	36.5–37.0	TCC	K	10900 ± 100	10440	10180	11900 ± 300	AA5433	WHG-662
KNR134-08 GGC38	38.2–38.7	TCC	L	16500 ± 120	16040	15780	18700 ± 170	AA4749	WHG-511
KNR134-08 GGC48	52.0–52.5	TOC	E	2380 ± 60	2320	1740	1700 ± 150	AA5934	WHG-885
KNR134-08 GGC48	90.2–90.7	TOC	G	3030 ± 50	2970	2390	2520 ± 150	AA5935	WHG-886
KNR134-08 GGC48	275.5–275.8	TOC	K	7450 ± 60	7390	6810	7640 ± 130	AA5936	WHG-887
KNR134-08 GGC48	283.8–284.3	TOC	L	7550 ± 70	7490	6910	7730 ± 130	AA5937	WHG-888
KNR134-08 GGC48	52.0–52.5	TCC	E	3940 ± 60	3480	3220	3510 ± 150	AA5939	WHG-875
KNR134-08 GGC48	90.2–90.7	TCC	G	7510 ± 70	7050	6790	7620 ± 130	AA5940	WHG-876
KNR134-08 GGC48	275.5–275.8	TCC	K	7560 ± 60	7100	6840	7660 ± 120	AA5941	WHG-877
KNR134-08 GGC48	283.8–284.3	TCC	L	13100 ± 100	12640	12380	14500 ± 230	AA5942	WHG-878
KNR134-08 GGC65	23.0–23.5	TOC	E	2810 ± 70	2750	2170	2200 ± 170	AA5879	WHG-854
KNR134-08 GGC65	51.0–51.5	TOC	G	3430 ± 60	3370	2790	2980 ± 160	AA5880	WHG-855
KNR135-08 GGC65	122.5–123.0	TOC	I	5080 ± 60	5020	4440	5120 ± 170	AA5882	WHG-857
KNR134-08 GGC65	155.2–155.7	TOC	K	7090 ± 100	7030	6450	7320 ± 140	AA5873	WHG-862
KNR134-08 GGC65	158.4–158.9	TOC	L	7230 ± 60	7170	6590	7440 ± 110	AA5874	WHG-863
KNR134-08 GGC65	23.0–23.5	TCC	E	7110 ± 80	6650	6390	7270 ± 120	AA5877	WHG-831
KNR134-08 GGC65	51.0–51.5	TCC	G	6300 ± 60	5840	5580	6400 ± 140	AA5878	WHG-835
KNR134-08 GGC65	155.2–155.7	TCC	K	7610 ± 60	7150	6890	7710 ± 130	AA5869	WHG-836
KNR134-08 GGC65	158.4–158.9	TCC	L	11500 ± 70	11040	10780	12700 ± 140	AA5870	WHG-837

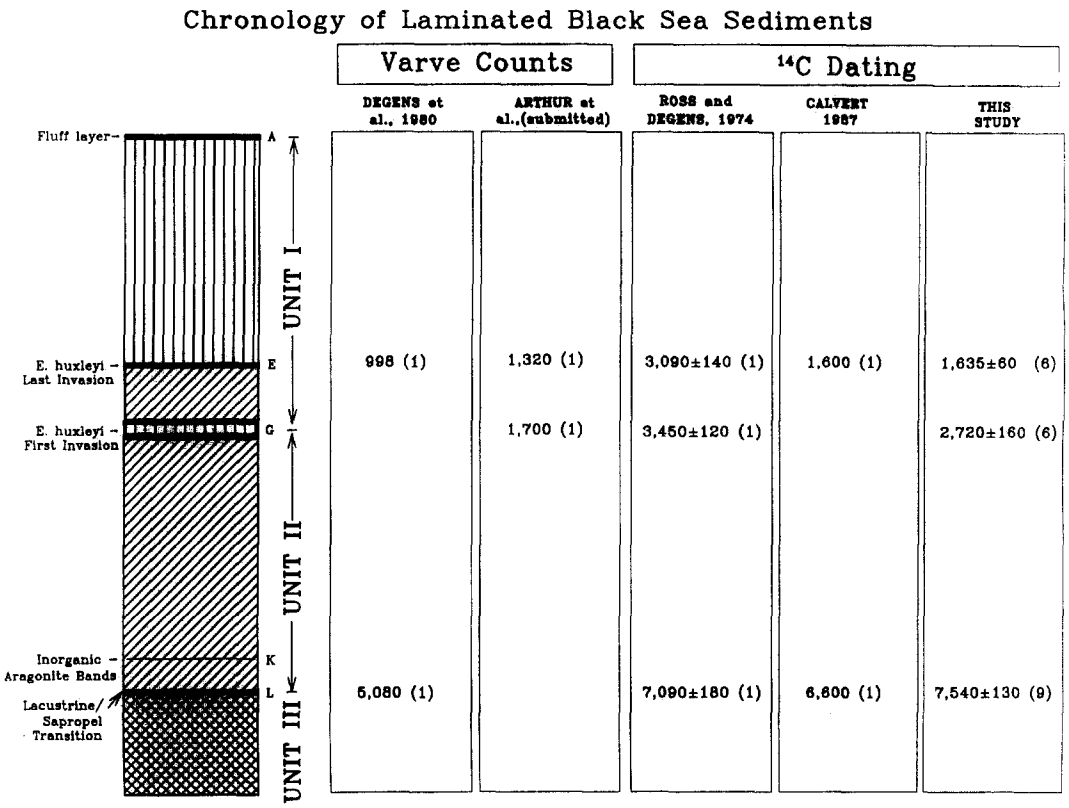

Fig. 7. Ages for the lithostratigraphic unit boundaries as reported by previous authors. Number of analyses used to estimate ages is given in parentheses after age.

GGC48 (205 cm), is 7560 ± 60 years BP and the organic carbon conventional radiocarbon age is 7450 ± 60 years BP. Applying the 460 and 60 year reservoir correction to these two fractions as determined for Unit I results in reservoir-corrected carbonate and organic carbon ages of 7100 and 7390, respectively. The 290 year difference is indistinguishable from the 320 years estimated for Unit I. Given that degree of consistency and the fact that the carbon flux in Unit I and II are similar (ARTHUR *et al.*, in press) we use a detrital organic carbon correction of 580 years for both Unit I and II sediments. The reservoir and detrital corrected radiocarbon age estimate of 6700 years BP for Horizon-L converts to a calendar age of 7540 years BP (STUIVER and BRAZIUNAS, 1993). This calendar age estimate for Horizon-L is significantly older than the varve-counted estimate of 5080 years BP (DEGENS *et al.*, 1980). Thus, the same possible errors in the varve-count ages as discussed earlier may also apply to Unit II sediments.

A brief comment on the history of anoxia

Several competing hypotheses have been put forth to explain the temporal history of anoxia in the Black Sea (e.g. DEUSER, 1974; GLENN and ARTHUR, 1985; CALVERT, 1987),

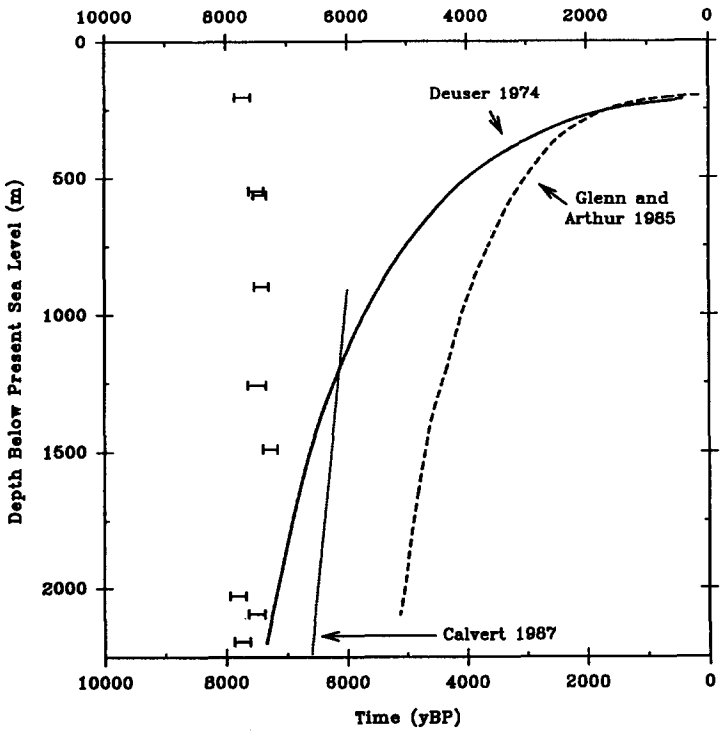

Fig. 8. Three models of Black Sea sapropel formation/evolution are compared to the total organic carbon ages (calendar years) for Horizon-L. The width of the symbol used (⊢⊣) reflects the 1-σ error in the age estimates for each measurement. This horizon marks the beginning of sapropel formation and the transition from oxygenated to anoxic conditions in the basin. None of the existing models fit the distribution of the data. Data suggests that all depths below at least 205 m went anoxic at the same time, rather than progressively shallowing over time.

yet none can fully explain the data presented (Fig. 8). DEUSER (1974) proposed that there was a time–depth evolution to anoxia with the deepest waters becoming anoxic at about 7300 years BP, or some 1700 years after post-glacial sea-level rise was thought by DEUSER (1974) to have reconnected the Mediterranean and Black Sea via the Bosporus Strait. The oxygen reservoir half-life was calculated to be approximately 1800 years and the oxic–anoxic interface rose through time to reach its present depth of 50–100 m. DEUSER (1974) further speculated that this interface will continue to rise approaching the 34 m sill depth of the Bosporus Strait asymptotically.

Using the varve chronology of DEGENS *et al.* (1980), GLENN and ARTHUR (1985) assumed that the connection with the Mediterranean occurred at around 7000 years BP and the first evidence of anoxia was at 5080 years BP in the deepest part of the basin. The oxygen reservoir half-life was calculated to be 1036 years, and the oxic–anoxic interface rose through time.

Additional radiocarbon data, corrected by the 2000 year varve-radiocarbon offset, on the E and L horizons from cores collected from 906 and 2238 m water depth led CALVERT (1987) to conclude that anoxia began at deep and mid-depths at approximately 6000 years

BP but sapropel formation ended (i.e. Horizon E) at 4000 years BP at mid-depth and 1000 years BP in the deep basin.

We have presented far more extensive chronological data than have been available to researchers previously. Our observation is that sapropel formation began at all depths (at least below 205 m) at approximately 7540 ± 130 years BP, which strongly suggests that the depth of the oxic–anoxic interface has not changed appreciably during the past 7540 years. This observation further suggests that the oxic–anoxic interface is controlled largely by the physical oceanography of the basin (e.g. sill depth, inflow/outflow rates, entrainment) and is not a depth evolving feature as speculated by DEUSER (1974), GLENN and ARTHUR (1985) or MURRAY *et al.* (1989). The data further show that Horizon-E was formed at the same time at all water depths, suggesting this lithologic feature is controlled by the initiation of increased *E. huxleyi* productivity and does not reflect a depth evolving change in water column chemistry as speculated by CALVERT (1987).

Our estimate that anoxia began at 7540 years BP is significantly older than the 5080 (varve) or even 7090 (uncorrected radiocarbon) years BP estimates proposed in earlier studies. Does this revised timing for the initiation of anoxia compare favorably with the timing of the reconnection of the Black Sea with the Mediterranean? The sill depth of the Bosporus Strait is 32–34 m (LATIF *et al.*, 1991). Using the radiocarbon dated coral records presented in FAIRBANKS (1990) to reconstruct the global sea-level curve for the past 17,000 years and then converting these ages to calendar ages (STUIVER and BRAZIUNAS, 1993) we find that eustatic sealevel was 34 m below the present level by 9800 years BP. We assume that the region around the Strait of Bosporus has been tectonically stable during the Holocene (MESCHERIKOV, 1971), and thus the initial post-glacial reconnection with the Mediterranean occurred at 9800 years BP. If true, our result suggests that 2300 years were required to consume all of the oxygen in the Black Sea at depths below the present oxic–anoxic interface. Assuming a deep-water oxygen value during the lacustrine phase of 300 μmol kg^{-1} (the approximate value for bottom waters (approximately 1600 m) in Lake Baikal, the world's deepest lake (WEISS *et al.*, 1991)) and the volume of the Black Sea below approximately 200 m water depth (495,000 km^3; DEUSER, 1974), we obtain an oxygen consumption of 6.6×10^{10} moles year^{-1}. This value requires a net flux of 1.9 g C m^2 year^{-1} across the oxic–anoxic interface if we make the extreme assumption that no new oxygen is supplied to the deep basin after 9800 years BP. If we make the other extreme assumption that oxygen was supplied to the deep basin at rates comparable to those of today for Mediterranean inflow and entrainment of surface waters (MURRAY *et al.*, 1991) and that no oxygen leaves the deep basin, then the flux of carbon across the oxic–anoxic interface would have been 9.7 g C m^2 year^{-1} to consume all of the oxygen within 2300 years. These carbon fluxes will be slightly larger if one incorporates the small fraction that was preserved in the sediments before the basin went anoxic. These two assumptions are extreme cases yet the values compare favorably with a carbon flux of 2.9 g C m^2 year^{-1} as measured in a sediment trap deployed at 250 m water depth (from HAY *et al.*, 1990) and with an estimate of net export of carbon across the oxic/anoxic interface of approximately 10.5 g C m^2 year^{-1} (KARL and KNAUER, 1991).

CONCLUSIONS

Several conclusions can be drawn from the data presented here. We disagree with MURRAY *et al.*'s (1991) argument that the pre-bomb Δ^{14}C of Black Sea surface waters were

depleted much more than the −50‰ normally assumed for surface waters of the world's oceans, with values as low as −200‰ per mil, equivalent to a reservoir-corrected age of 1430 years. They argue that values this low explain the apparently old radiocarbon ages from Unit I sediments as compared to the varve counting age estimates. Our data on a mollusc collected live in 1931 reveal the pre-bomb $\Delta^{14}C$ surface water values were −55‰ (460 years).

Radiocarbon analyses of Unit I sediments from three box cores recovered from the east, central, and western sectors of the central Black Sea reveal within-core and core-to-core consistencies in both the organic carbon and carbonate sediment fractions. These results suggest that the maximum "detrital" correction that can be applied to these sediments is 580 years for the organic carbon and 260 years for the carbonate fraction, and not 2000 years as has been assumed previously (e.g. DEGENS *et al.*, 1980; HAY *et al.*, 1991).

The age of Horizon-E or Unit 1/2 boundary of Ross and DEGENS (1974) and Horizon G or the first invasion of *E. huxleyi* by HAY *et al.* (1991) has been determined to be 1635 ± 60 and 2720 ± 160 years BP respectively in three cores. These age estimates are made from both the organic carbon and carbonate fractions, includes a reservoir correction, detrital carbon correction and have been converted to calendar years to better compare with the varve chronology of DEGENS *et al.* (1980), HAY *et al.* (1991) and ARTHUR *et al.* (in press). The most recent estimate for these boundaries from varve counting comes from ARTHUR *et al.* (in press) and have been found to be 1320 and 1700 years, respectively. These estimates are 20 and 35% less than the radiocarbon-derived chronology presented here and compares favorably with recent sediment trap data suggesting varves would have been formed in only 4 of the 5 years that trap data was available. These data suggest that the past discrepancies between the varve and radiocarbon chronologies have been resolved.

Radiocarbon ages of the organic carbon and carbonate fractions from Unit II sediments reveal that the best estimate for the age of Horizon-L is 7540 ± 130 years BP calendar years. This estimate contrasts with the varve counting estimate of 5080 years for this boundary.

The radiocarbon results of Horizon-L sediments from cores recovered from water depths spanning the range 205–2195 m reveals that the entire basin went anoxic at the same time. The level of the present oxic/anoxic boundary seems to have been stable for the past 7500 years, is controlled by the physical oceanography of the basin, and has not evolved depthwise over time as assumed by previous workers (e.g. DEUSER, 1974; GLENN and ARTHUR, 1985; CALVERT, 1987; MURRAY *et al.*, 1989).

The Black Sea was reconnected with the Mediterranean Sea via the Bosporus Strait at approximately 9800 years BP and 2300 years were required to consume all of the oxygen in the basin below approximately 200 m water depth. A carbon flux into the deep basin of 1.9 to 9.7 g C m^2 $year^{-1}$ is required, and is consistent with estimates of the modern carbon flux across the oxic/anoxic interface, suggesting relatively stable carbon production in the Black Sea throughout much of the Holocene.

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