Aegean Sea as driver of hydrographic and ecological changes in the eastern Mediterranean

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ABSTRACT

The eastern Mediterranean is undergoing a long-term increase in net evaporation, which may have preconditioned the profound changes that occurred in its deep-sea ventilation over the past two decades. We test the sensitivity of Aegean convective deep-water formation to forcing in the opposite sense, based on a last interglacial episode of enhanced freshwater injection into the eastern Mediterranean. We find that Aegean subsurface ventilation collapsed completely within 40 ± 20 yr, promoting euxinic conditions hostile to aerobic life that expanded toward the photic layer within 650 ± 250 yr. Similar conditions extended throughout the eastern Mediterranean 300 ± 120 yr later. These findings emphasize the exceptional sensitivity of Aegean deep-water formation to climate forcing, driving large-scale hydrographic adjustments throughout the eastern Mediterranean and beyond.

Keywords: Mediterranean Sea, thermohaline circulation, sapropels, stable isotopes, biomarkers.

INTRODUCTION

On a global scale, new deep-water formation delivers oxygen to the deep sea. In the eastern Mediterranean, this process has long been dominated by the sinking and spreading of Adriatic dense waters (Malanotte-Rizzoli and Hecht, 1988). In the late 1980s–early 1990s, there was an abrupt shift to dominance of Aegean deep waters in the eastern Mediterranean deep-sea ventilation (Eastern Mediterranean Transient) (Roether et al., 1996; Klein et al., 1999; Malanotte-Rizzi et al., 1999) that eventually influenced the density of Mediterranean outflow into the Atlantic (Millot et al., 2006). Several studies conclude that this profound hydrographic reorganization was preconditioned by a long-term increase in eastern Mediterranean net evaporation (Béthoux et al., 1998; Boscolo and Bryden, 2001; Skliris and Lascaratos, 2004; Skliris et al., 2007). Other studies emphasize the role of synoptic changes in the wind field affecting upper thermocline circulation and, in turn, the salt supply to the Aegean Sea (Malanotte-Rizzi et al., 1999; Samuel et al., 1999; Stratford and Haines, 2002).

We investigate the sensitivity of the Aegean thermohaline circulation to a reduction of eastern Mediterranean net evaporation due to freshwater flooding, using a key example of the organic-rich layers (sapropels) that periodically occur in the eastern Mediterranean sedimentary archive. Sapropels reflect periods of sluggish bottom-water ventilation in response to enhanced monsoon-fueled river discharge along the North African margin (e.g., Rohling et al., 2002; Emeis et al., 2003). Last interglacial sapropel S5 is intensely developed and holds excellent potential for high-resolution studies (Cane et al., 2002; Rohling et al., 2002, 2004, 2006). However, a lack of Aegean S5 records has, to date, limited our understanding of the hydrographic responses in this critical region to a sharp reduction in eastern Mediterranean net evaporation.

Here we present the first systematic high-resolution multiproxy study of an Aegean S5, as retrieved from southeastern Aegean core LC21 (Fig. 1). Results are discussed within the context of previously described contemporaneous records from the open eastern Mediterranean Ocean Drilling Program (ODP) Site 971A (Cane et al., 2002; Rohling et al., 2002, 2004, 2006).

MATERIAL AND METHODS

Core LC21 was recovered in 1995 by RV Marion Dufresne for the EC-MAST2 (Marine, Science, and Technology Programme) PALAEO-FLUX (Biogeochemical Fluxes in the Mediterranean Water-Sediment System) program. We analyzed S5, in sections 5 and 6, at 1 cm (decadal) resolution for δ18O and δ13C in Globigerinoides ruber (white) and Neogloboquadrina pachyderma (dextral), and at 5 cm (centennial) resolution for alkene-based sea surface temperatures (SSTs), total organic carbon (Corg), and isorenieratene concentrations (GSA Data Repository Fig. DR1).

Stable isotope analyses were performed in Southampton with a Europa Geo2020 dual inlet mass spectrometer following individual acid–bath reaction of 15–20 handpicked and cleaned specimens in the size range 300–350 μm (G. ruber) and 250–300 μm (N. pachyderma). Isotope ratios are expressed as δ18O and δ13C, in permil relative to Vienna PeeDee belemnite (VPDB). External precision was better than 0.06‰ for both δ13C and δ18O.

The Corg contents, alkene analyses, and isorenieratene concentrations were performed at the Royal Netherlands Institute for Sea Research. The Corg contents were determined on decalcified sediments, using a Carlo Erba Flash elemental analyzer coupled to a Thermofinnigan DeltaPLUS isothermal remanent magnetization mass spectrometer (MS) system. For alkene analysis, freeze-dried and homogenized sediments were extracted using the Dionex accelerated solvent extraction (ASE) technique using dichloromethane (DCM)/methanol (2:1, v/v) at 100 °C and 7.6 × 109 Pa. The extracts were separated by Al2O3 column chromatography.

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raphy using first hexane/DCM (9:1, v/v) and then hexane/DCM (1:1, v/v) to elute the alkenone fraction, which was analyzed by gas chromatography (GC) and GC/MS. Alkenone-based SSTs were calculated following Müller et al. (1998). Isorenieratene concentrations were determined following Hopmans et al. (2005).

RESULTS AND DISCUSSION

Sapropel S5 in LC21 is the most intensely developed sapropel known from the Pleistocene, with \( C_{\text{org}} \) concentrations as high as 14% and a thickness of ~120 cm (Fig. DR1), deposited over a period of ~5 k.y. (e.g., Rohling et al., 2002). The absence of benthic fossils, the preserved sedimentary laminations in several intervals, and the occurrence of specific biomarkers (see following) together indicate euxinic conditions in the Aegean water column during S5 deposition. In the absence of bioturbation, the great thickness of S5 in LC21 allows very high sampling resolution.

We compare our results for S5 from Aegean core LC21 with those from open eastern Mediterranean ODP Site 971A (Fig. 1). Using the statistical multiproxy correlation approach for records through S5 (Cane et al., 2002; Rohling et al., 2002), the LC21 record has been transferred onto the 971A-equivalent depth scale that serves as a master depth scale for S5 in the open eastern Mediterranean (Appendix DR1 and Fig. DR1). This method allows a direct comparison between signals recorded in Aegean core LC21 and those in open eastern Mediterranean ODP Site 971A, with a vertical uncertainty of less than ±1 cm (1σ = ±0.83 cm) within S5 on the 971A-equivalent depth scale. This correlation indicates an exact coincidence of the large shift to light \( \delta^{18}O_{\text{rular}} \) near the onset of S5 in the two cores (Fig. 2B). Given the basin’s prevalent wind-driven surface circulation and the proximity of the two sites (<300 km), such a large and abrupt signal is expected to be synchronous within the 40 ± 20 yr temporal resolution of our record. We are therefore confident that our statistical method delimits the correlations well within the 1σ bounds for this crucial segment of the records.

Our chronology for S5 relies on a simple linear interpolation between the estimated ages of the onset and termination of the associated light \( \delta^{18}O_{\text{rular}} \) anomaly (Rohling et al., 2002), suggesting that, within S5, 1 cm corresponds to ~40 ± 20 yr in LC21 and ~200 ± 80 yr in 971A (Appendix DR1). The sediment mass accumulation rate (SMAR) in LC21 exceeds that in 971A by 5 times. Given this SMAR difference and that \( C_{\text{org}} \) concentrations reach 14% in LC21 compared to 7% in 971A, it appears that the \( C_{\text{org}} \) burial flux during S5 deposition was a full order of magnitude higher in the Aegean Sea than in the open eastern Mediterranean.

Figure 2 shows the signals through S5 in LC21 along with their counterparts in ODP Site 971A (all data on the common 971A depth scale). At both sites, the absolute \( \delta^{18}O_{\text{pachyderma}} \) values are virtually identical (Fig. 2C). This supports the previous notion that the isotopic composition of \( N. \text{pachyderma} \) reflects basin-integrated property changes, suggesting a habitat in (top) intermediate waters deriving from a single source region (Rohling et al., 2004). In Aegean core LC21, changes in \( \delta^{18}O_{\text{rular}} \) appear to closely track \( \delta^{18}O_{\text{pachyderma}} \) within S5, which contrasts with strong variable offsets between \( \delta^{18}O_{\text{rular}} \) and \( \delta^{18}O_{\text{pachyderma}} \) in 971A (Fig. DR2). The \( \delta^{18}O_{\text{rular}} \) represents near-surface conditions in the summer mixed layer (Rohling et al., 2004), and the similarity between \( \delta^{18}O_{\text{rular}} \) and \( \delta^{18}O_{\text{pachyderma}} \) in LC21 suggests that both species inhabited water masses with similar properties. This would agree with modern observations that the southeastern Aegean site of LC21 is directly in the northward flow path of Levantine surface water (Theocharis et al., 2002), which derives from the area where Levantine intermediate water is formed, and that the intermediate water reaches depths as shallow as 70 m in the Aegean due to prevailing northerly winds and the basin’s general cyclonic circulation (Poulos et al., 1997; Pinardi and Masetti, 2000; Zervakis et al., 2004). Work on the Holocene period of S1 deposition indicates similar conditions during times of decreased eastern Mediterranean net evaporation (Myers et al., 1998; Casford et al., 2002).

Given that the \( \delta^{18}O_{\text{pachyderma}} \) records through S5 are similar for both sites, the contrast between \( \delta^{18}O_{\text{rular}} \) and \( \delta^{18}O_{\text{pachyderma}} \) in 971A can be entirely attributed to variability in \( \delta^{18}O_{\text{rular}} \). The \( \delta^{18}O_{\text{rular}} \) signal in 971A displays lighter values (the offset from \( \delta^{18}O_{\text{pachyderma}} \) reaches more than 1.5‰; Fig. DR2) and also is more variable than in LC21 (Fig. 2B). Because both sites have similar SSTS (Fig. 2A), this difference in \( \delta^{18}O_{\text{rular}} \) is unlikely to be attributable to SST contrasts. The \( \delta^{18}O_{\text{rular}} \) differences within S5 contrast with the similarity of \( \delta^{18}O_{\text{rular}} \) at the two sites before and after S5. The observed differences between the \( \delta^{18}O_{\text{rular}} \) signals within S5, along with similarity of \( \delta^{18}O_{\text{pachyderma}} \) and SST signals in LC21 and 971A as well as other sites throughout the eastern Mediterranean (Rohling et al., 2002), may only be explained by limited interaction between surface and intermediate waters at the location of Site 971A, which is remote from the intermediate-water source region.

Spatially, \( \delta^{18}O_{\text{rular}} \) is lighter around the location of 971A than elsewhere in the open eastern Mediterranean (Rohling et al., 2002), as observed for other sapropels (Rohling and De Rijk, 1999; Emeis et al., 2003). This pattern has been ascribed to extensive (monsoon fueled) freshwater drainage along the wider North African margin, through currently dry river (wadi) systems, supplementing—but more variable than—Nile River discharge (Rohling et al., 2002, 2004; Scrivner et al., 2004). The \( \delta^{18}O_{\text{rular}} \) values in the brief interval of reduced discharge (Rohling et al., 2002) from the wider North African margin (56–52 cm in 971A) decrease back to the typical \( \delta^{18}O_{\text{rular}} \) Values found throughout S5 in LC21 (Fig. 2B). This suggests that the Aegean record reflects an underlying \( \delta^{18}O_{\text{rular}} \), controlled by less intense freshwater dilution that may reflect propagation through the basin of the surface freshening caused by North African river input into the open eastern Mediterranean (Rohling et al., 2002; Scrivner

Figure 1. Map of Mediterranean showing main patterns of surface circulation (gray arrows), sites of eastern Mediterranean dense water formation (shaded areas) (Pinardi and Masetti, 2000), and locations (black dots) of core LC21 (35° 40′N, 26°35′E; 1522 m water depth) and Ocean Drilling Program Site 971A (33°43′N; 24°41′E; 2026 m water depth). Aeg.—Aegean; Lev.—Levantine; Ad.—Adriatic; Ion.—Ionian.
Superimposed on this widespread average mixed signal, the large regional δ¹⁸Oₘₗₜ anomalies found in the open eastern Mediterranean (notably near 971A) would reflect more direct impacts of the river-borne monsoon floods.

LC21 and 971A simultaneously reach the lightest δ¹⁸Oᵣₘ values ~650 ± 250 yr after the sharp δ¹⁸Oᵣᵢₗ shift that marks the onset of freshwater flooding into the basin (Fig. 2B). This peak in eastern Mediterranean sea-surface freshening coincides with the appearance of very light δ¹³Cₚₙ₅₃₃ₕ₅₃₃ values (lighter by ~5‰ than δ¹³Cᵣₑₚ₅; Fig. DR2) at both sites, following ~400 yr of near absence of this species throughout the eastern Mediterranean (Rohling et al., 2006). The shift to light δ¹³Cₚₙ₅₃₃ₕ₅₃₃ values, observed in both records, has been suggested to reflect the proximity of a subsurface reservoir of isotopically light metabolized carbon to the subsurface habitat of *N. pachyderma* (Rohling et al., 2006). At the same level in LC21, high concentrations appear of isorenieratene (>9 µg/g sediment), a specific aromatic carotenoid of anaerobic, photolithotrophic green sulfur bacteria (Chlorobiaceae) (Fig. 2G). These bacteria require both sulfide and light (Repeta et al., 1989; Koopmans et al., 1996; Passier et al., 1999). Similar to the reconstruction for the open eastern Mediterranean (Rohling et al., 2006), the abrupt appearance of high isorenieratene concentrations in the Aegean Sea, combined with the occurrence of subthermocline foraminiferal species (e.g., *N. pachyderma*), reflects the development of euxinic conditions throughout the Aegean water column up to ~200 m. Comparison of LC21 with 971A (Fig. 2G) illustrates that such conditions had developed in the Aegean Sea 100 ± 40 to 300 ± 120 yr before they became established in the open eastern Mediterranean.

A clear sequence of causes and effects can now be summarized for the last interglacial episode of freshwater flooding into the eastern Mediterranean. Given that benthic aoxic conditions and a sharp Cₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑₑᵉ⁾ anomalously, our findings for the 55 period highlight an exceptional sensitivity of the Aegean to changes in wider eastern Mediterranean climate forcing of any sign, with responses that lead to profound hydrographic adjustments that subsequently propagate throughout the open eastern Mediterranean, and that potentially affect even the North Atlantic (Millot et al., 2004).
et al., 2006). Both increases (preconditioning modern changes) and decreases (this study) in eastern Mediterranean net evaporation are found to drive basin-scale reorganizations in subsurface water-mass dynamics, with extensive ecological impacts.

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Appendix DR1: Supplementary Information on stratigraphic framework and chronology

Stratigraphic framework
The approach we use to build a stratigraphic framework for core LC21 statistically seeks the best correlation with ODP core 971A, which was previously proposed as “master record” for S5 in the eastern Mediterranean (eMed) (Cane et al., 2002). In this correlation none of the correlation markers were a priori assumed to be syn-/diachronous. Distinct and unambiguously defined stratigraphic markers based on five independent proxy records have been recognized on a basin scale during the S5 deposition (Figure DR1, panel II). Remarkable O and C isotope shifts, clear faunal presence/absence and sharp peaks in the C\textsubscript{org} and isorenieratene abundance have been recognized in Aegean LC21 and western Levantine 971A (Figure DR1, Table DR1). Markers, such as faunal zero abundance levels of the most abundant foraminiferal species, and sharp peaks in the isotopic profiles have been assigned a primary role, and these were used to establish the correlation between the two sites. Markers deriving from less well-defined events, and shifts in organic carbon content and isorenieratene abundance have been assigned a secondary role and were used for validation of the regression only. In order to avoid possible bias caused by post-depositional oxidation, the visual extent of the dark-coloured sapropel sediments was not used to identify the calibration (Cane et al., 2002).

In Figure DR1 (panel II), 11 primary (grey open diamonds) and the 12 secondary (black solid circles) correlation marker pairs for LC21 and 971A are plotted against one-another. Linear regression between primary correlation markers is highly significant ($R^2$=0.97). Moreover, the regression is corroborated by the proximity of the secondary markers to the linear fit. Nevertheless, a second-order polynomial regression between primary
correlation markers ($R^2=0.996$), albeit statistically indistinct from the linear regression, yields a visual “point to point” improvement. Therefore, the latter regression has been employed to calibrate the LC21 depth scale to 971A-equivalent depths. The 1σ uncertainty that applies to the correlation based on the second-order polynomial fit through the primary correlation markers equals ±0.83 cm.

**Chronology**

The results presented here support the concept of a non-synchronous inception of anoxic conditions throughout the basin during the last interglacial period (Cane et al., 2002). In contrast, the onset of the monsoon-fuelled freshwater dilution of the eMed surface waters has been virtually synchronous throughout the basin, preceding the organic-rich sedimentation (Rohling et al., 2002a). Since the eMed sea surface is the principal source of moisture for precipitation over Soreq Cave (N. Israel) (Matthews et al., 2000), any appreciable change in the oxygen isotopic composition of eMed sea surface waters (as reflected in $\delta^{18}O_{rubr}$) will be recorded directly by a comparable shift in the $\delta^{18}O_{calcite}$ of Soreq Cave speleothems (Bar-Matthews et al., 2000). Therefore, in agreement with Rohling et al. (2002a), we contend that the onset of the humid phase in the eMed region during the Marine Isotopic Stage 5e (MIS 5e) is synchronous at a basin-scale and can be assigned an approximate age of 124 ka B.P. (Bar-Matthews et al., 2000). Similarly, the major return of $\delta^{18}O_{rubr}$ to pre-sapropel values near the end of S5 deposition is synchronous between LC21 and 971A and throughout the eMed (Rohling et al., 2002a), and matches the termination of the humid phase in Soreq Cave dated at around 119 ka B.P. (Bar-Matthews et al., 2000).

The chronology for S5 provided in the present study relies on a simple linear interpolation (i.e. assuming a constant sedimentation rate throughout the S5) between the estimated ages of the onset (124 ka B.P.) and the termination (119 ka B.P.) of the depletion and enrichment trends in $\delta^{18}O_{rubr}$. On the basis of this very simple calculation, and taking into account a 2 k.y. uncertainty in the duration of the humid period in Soreq Cave (Bar-Matthews et al., 2000), 1 cm corresponds to $39.7\pm20$ years in core LC21, and $199.4\pm80$ years in core 971A. An analogous coincidence between major shifts in
\(\delta^{18}O_{\text{calcite}}\) of the Soreq Cave speleothems and in \(\delta^{18}O_{\text{rubber}}\) in LC21 (Rohling et al., 2002b) is associated with a widespread humid event at the time of deposition of early- to mid-Holocene sapropel S1 (see also Arz et al., 2003). The timing relationship between LC21 and Soreq Cave during the early- to mid-Holocene is supported by independent datings performed on both archives (Bar-Matthews et al., 2000; Mercone et al., 2000), and the close agreement between those results corroborates our approach of using the datings from Soreq Cave to provide a chronology for sapropel S5.

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Figure DR1. Left I) Down-core records of different geochemical proxies through S5 for core LC21. Shaded area represents the visual extent of the dark-coloured sapropel S5 sediments. It corresponds to a partly laminated, benthic aozic interval indicating persistent sea-floor anoxia. A: $\delta^{18}O$ records for the summer mixed layer dwelling *Globigerinoides ruber* (white) (grey dashed line) and top intermediate water dwelling *Neogloboquadrina pachyderma* (dextral) (black dashed line) planktonic foraminifera. B: $\delta^{13}C$ records for *G. ruber* (white) and *N. pachyderma*. The isotopic records ($\delta^{18}O$ and $\delta^{13}C$) of *G. ruber* (grey solid line) and *N. pachyderma* (black solid line) were lowess smoothed (tension 0.04 and 0.05, respectively) in order to highlight main trends. C: SST reconstructions based on the long-chain alkenone unsaturation index (black solid line). D: Organic carbon content (Corg, wt%) (black filled area). E: Isorenieratene abundance calculated with respect to the total amount of sediment (grey filled area). Right II) Linear (grey line) and polynomial (black line) fits through the primary correlation markers (grey open diamonds), using ODP 971A as the independent variable (“bench mark”) and LC21 as dependent variable. Black open circles represent secondary correlation markers. Big + and grey blocks indicate the visual extent of the dark-coloured sapropel sediments. The time scale on the right is based on the correlation between major shifts in the $\delta^{18}O_{rubber}$ at sapropel onset and termination and the onset and termination of the humid period in Soreq Cave (Rohling et al., 2002a).
Figure DR2. A: alkenone-based SST (open circles), $^{18}\text{O}_{\text{pachyderma}}$ (black line), and $^{18}\text{O}_{\text{rub}}$ (grey line) records in 971A; B: alkenone-based SST (open circles), $^{18}\text{O}_{\text{pachyderma}}$ (black line), and $^{18}\text{O}_{\text{rub}}$ (grey line) records in LC21; C: $^{13}\text{C}_{\text{pachyderma}}$ (black line), and $^{13}\text{C}_{\text{rub}}$ (grey line) records in 971A; D: $^{13}\text{C}_{\text{pachyderma}}$ (black line), and $^{13}\text{C}_{\text{rub}}$ (grey line) records in LC21. Labels identify correlation markers (Table DR1).
<table>
<thead>
<tr>
<th>Correlation point</th>
<th>Species/stable isotope/org. geochem.</th>
<th>Definition</th>
<th>Confidence</th>
<th>971A Depth</th>
<th>LC-21 Depth</th>
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<td><em>N. pachyderma</em> δ¹³C</td>
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<td>39.25</td>
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<td>prominent isotopic depletion peak after sapropel onset</td>
<td>secondary</td>
<td>59.25</td>
<td>990.00</td>
</tr>
<tr>
<td>isor1</td>
<td>isorenieratene org. geochem.</td>
<td>last peak in isorenieratene abundance</td>
<td>secondary</td>
<td>43.50</td>
<td>895.00</td>
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<tr>
<td>isor2</td>
<td>isorenieratene org. geochem.</td>
<td>Highest value in isorenieratene abundance after the sapropel onset</td>
<td>secondary</td>
<td>56.50</td>
<td>980.0</td>
</tr>
<tr>
<td>isor3</td>
<td>isorenieratene org. geochem.</td>
<td>sample before preceding isorenieratene occurrence</td>
<td>secondary</td>
<td>58.50</td>
<td>990.0</td>
</tr>
<tr>
<td>C&lt;sub&gt;org&lt;/sub&gt;1</td>
<td>C&lt;sub&gt;org&lt;/sub&gt; org. geochem.</td>
<td>last sample at the sapropel termination where C&lt;sub&gt;org&lt;/sub&gt;&lt;sub&gt;&lt;&lt;/sub&gt;2%</td>
<td>secondary</td>
<td>40.50</td>
<td>885.00</td>
</tr>
<tr>
<td>C&lt;sub&gt;org&lt;/sub&gt;2</td>
<td>C&lt;sub&gt;org&lt;/sub&gt; org. geochem.</td>
<td>peak in C&lt;sub&gt;org&lt;/sub&gt; after sapropel onset</td>
<td>secondary</td>
<td>62.50</td>
<td>1007.0</td>
</tr>
<tr>
<td>C&lt;sub&gt;org&lt;/sub&gt;3</td>
<td>C&lt;sub&gt;org&lt;/sub&gt; org. geochem.</td>
<td>last sample before C&lt;sub&gt;org&lt;/sub&gt;&gt;1% at the sapropel onset</td>
<td>secondary</td>
<td>56.50</td>
<td>985.00</td>
</tr>
<tr>
<td>top black</td>
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<td>40.25</td>
<td>885.00</td>
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<tr>
<td>base black</td>
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<td>63.00</td>
<td>1005.0</td>
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