



Seasonal hypoxia was a natural feature of the coastal zone in the Little Belt, Denmark, during the past 8 ka



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ABSTRACT

The extent of the hypoxic area in the Baltic Sea has rapidly expanded over the past century. Two previous phases of widespread hypoxia, coinciding with the Holocene Thermal Maximum (HTM; 8–4 ka before present; BP) and the Medieval Climate Anomaly (MCA; 2–0.8 ka BP), have been identified. Relatively little is known about bottom water redox conditions in the coastal zone of the Baltic Sea during the Holocene, however. Here we studied the geochemical composition of a sediment sequence from a currently seasonally hypoxic site in the Danish coastal zone, the Little Belt, retrieved during Integrated Ocean Drilling Program Expedition 347 (Site M0059). The base of the studied sediment sequence consists of clays low in organic carbon (C_{org}), molybdenum (Mo) and iron sulfides (Fe-sulfides), and rich in iron oxides (Fe-oxides), indicative of a well-oxygenated, oligotrophic (glacial) meltwater lake. An erosional unconformity separates the glacial lake sediments from sediments that are rich in C_{org} . The absence of Mo, in combination with high C_{org}/S values, indicates that these sediments were deposited in a highly productive, well-oxygenated freshwater lake. The transition to modern brackish/marine conditions was very rapid, and subsequent continuous sequestration of Mo in the sediment and high ratios of reactive iron (Fe_{HR}) over total Fe (Fe_{TOT}) suggest (seasonal) hypoxia occurred over the last ~8 ka. Maxima in sediment C_{org} , Mo and Fe_{HR}/Fe_{TOT} ratios during the HTM and MCA suggest that the hypoxia intensified. Our results demonstrate that the Little Belt is naturally susceptible to the development of seasonal hypoxia. While periods of climatic warming led to increased deoxygenation of bottom waters, high nutrient availability in combination with density stratification were likely the main drivers of hypoxia in this part of the coastal zone of the Baltic Sea during the Holocene.

1. Introduction

Dissolved oxygen concentrations have been decreasing in many parts of the world's oceans over the past decades (e.g. Whitney et al., 2007; Stramma et al., 2008; Karstensen et al., 2015). Particularly coastal bottom waters are increasingly suffering from hypoxia, i.e. dissolved oxygen concentrations < 2 mg/L (Diaz and Rosenberg, 2008). The expansion of coastal hypoxia has led to a decreasing diversity of benthic communities, fish habitat loss and the development of benthic dead zones (e.g. Rabalais et al., 2002; Diaz and Rosenberg, 2008; Vaquer-Sunyer and Duarte, 2008). Increased anthropogenic nutrient input is the primary cause of deoxygenation in coastal waters

(e.g. Diaz and Rosenberg, 2008), which can be further enhanced by global warming through its impact on gas solubility, water column ventilation and enhanced marine primary productivity (e.g. Keeling et al., 2010; Carstensen et al., 2014a; Hallegraeff, 2010).

The Baltic Sea is a prime example of a basin where bottom water hypoxia has expanded due to human activities (Gustafsson et al., 2012) and has been amplified due to climatic warming (Kabel et al., 2012; Carstensen et al., 2014a). Over the past century, the hypoxic area in the Baltic Sea has increased tenfold (Carstensen et al., 2014a), consequently leading to major changes in the abundance and spatial distribution of benthic fauna (Karlson et al., 2002; Conley et al., 2009). The Baltic Sea is particularly sensitive to hypoxia because of its estuarine circulation

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and its permanent halocline at depths between 40 and 80 m in the deeper central parts. As a consequence of the circulation pattern and the strong density stratification, vertical mixing is limited and water in the basin has a relatively long residence time of ~30 years (e.g. Wulff et al., 1990; Stigebrandt and Gustafsson, 2003). In combination with the excessive anthropogenic nutrient load over the past century, the aforementioned stratification has caused the Baltic Sea to become highly eutrophic (e.g. Gustafsson et al., 2012).

During the Holocene at least two other phases of widespread hypoxia have been recognized in the Baltic Sea (Zillén et al., 2008). The postglacial development of the Baltic Sea is characterized by an alternation between freshwater lake and brackish/marine conditions, which resulted from the complex interplay of the retreating ice sheet, isostatic rebound and eustatic sea level variations (e.g. Björck, 1995; Sohlenius et al., 1996; Andrén et al., 2011). Around 8 ka before present (BP) a permanent open connection with the North Sea was gradually established, leading to the modern brackish/marine conditions. The full transition from freshwater to brackish/marine conditions is observed in sedimentary records throughout the Baltic Sea. It is characterized by the widespread occurrence of laminated sediments (e.g. Zillén et al., 2008), a marked increase in sedimentary organic carbon content (C_{org} ; e.g. Sohlenius et al., 2001), enrichments in molybdenum (Mo; e.g. Jilbert and Slomp, 2013; Dijkstra et al., 2016), the occurrence of the Fe-sulfide mineral greigite (Fe_3S_4 ; Loughheed et al., 2012; Reinholdsson et al., 2013) and the replacement of freshwater species of, for example, diatoms and dinoflagellates, by brackish/marine species (e.g. Andrén et al., 2000a; Brenner, 2005). Laminated, C_{org} , Mo and greigite rich sediments are deposited and preserved in hypoxic settings. The hypoxic interval from 8 to 4 ka BP (Zillén et al., 2008), coincides with the Holocene Thermal Maximum (HTM; ~9–5 ka BP; Snowball et al., 2004; Renssen et al., 2012) a period of relatively warmer climate. At the same time (~7–4 ka BP) the salinity in the Baltic Sea was at its maximum (e.g., Berglund, 1971; Gustafsson and Westman, 2002). The coupling of the warming and the establishment of a permanent halocline associated with marine water ingress are thought to have led to the oxygen depletion (Zillén et al., 2008). A second interval with widespread occurrence of laminated sediments, and a marked increase in sedimentary organic carbon content and Mo (e.g. Jilbert and Slomp, 2013; Dijkstra et al., 2016), indicative of hypoxic conditions, occurred between 2 and 0.8 ka BP (Zillén et al., 2008). This hypoxic interval also coincides with a period of regional climate warming: the Medieval Climate Anomaly (MCA; e.g. Esper et al., 2002). Furthermore, during this period the human population in the drainage area of the Baltic Sea rapidly increased, leading to large scale agricultural land use and soil nutrient release (Zillén and Conley, 2010). Together with regional climate warming, human activities thus may have driven the development of this second phase of widespread hypoxia (e.g. Carstensen et al., 2014b).

While the modern expansion of hypoxia has affected both the deep basins and the coastal zone of the Baltic Sea (Conley et al., 2011), relatively little is known about the bottom water redox conditions in the coastal zone of the Baltic Sea during the Holocene. On the one hand, the availability of nutrients is much higher in coastal waters, making these waters more vulnerable to eutrophication (e.g. Diaz and Rosenberg, 2008). On the other hand, coastal regions are often shallower, allowing better mixing of the water column, and therefore less stratification. The complex interplay of the factors leading to oxygen depletion in combination with the highly dynamic nature of the coastal zone makes it hard to reconstruct its past and predict its future redox state. Recently, Ning et al. (2016) showed that Gås fjärden (a coastal site in southeast Sweden, that is presently intermittently hypoxic in late summer) was continually hypoxic before 3 ka BP. This suggests that hypoxia in the coastal zone was more persistent in the past, when three effects were occurring simultaneously: (1) the water depth at today's coastal sites was deeper due to local isostasy; (2) the Baltic Sea halocline was shallower due to more marine water ingress and

higher salinity; (3) climate was warmer due to the HTM. Ning et al. (2016) note that the Gås fjärden area was not hypoxic during the MCA period, which they attributed to the lack of a halocline and a shallower water depth resulting from isostatic rebound. These complex interactions illustrate that the drivers of hypoxia, such as changes in climate, nutrient input, local water depth and salinity, may be different in coastal and offshore areas.

Here we studied a Holocene sediment sequence from the south-eastern Danish coastal zone, located in the Little Belt (Site M0059), which was retrieved during Integrated Ocean Drilling Program (IODP) Expedition 347. The high sedimentation rate at this location (~50 m sediment deposition in the past ~8 ka; Andrén et al., 2015), which has been seasonally hypoxic since the 1970s (Karlsén et al., 2002; Conley et al., 2007), and its proximity to the connection with the North Sea, make this site suitable for a detailed study of the freshwater lake to brackish/marine transition and the evolution of bottom water redox conditions throughout the Holocene. A variety of inorganic geochemical records, including bulk elemental data, as well as sulfur and iron speciation data, combined with a ^{14}C -based age model, shows that the transition to brackish/marine conditions was rapid and led to the development of persistent (seasonal) hypoxia.

2. Background

The Baltic Sea (Fig. 1a) was formed approximately 16 to 15 ka before present (BP) right after the last glaciation (e.g. Houmark-Nielsen and Kjaer, 2003). The retreating ice sheet left behind a large meltwater-filled ice lake, generally referred to as the Baltic Ice Lake (e.g. Björck, 1995). Around the onset of the Holocene (11.7 ka BP; Walker et al., 2009), the Baltic Ice Lake drained down to sea level through an outlet in south central Sweden (Jakobsson et al., 2007) and a new stage, the Yoldia Sea stage, began.

During the following ~350 years, freshwater from the melting ice sheet was still flowing out of the Baltic basin preventing any marine water to enter the basin. A small climate deterioration, the Preboreal oscillation, reduced the melt water flux and marine waters entered the basin around 11.3 ka BP. This brief and relatively weak brackish phase only lasted for a maximum of 350 years after which the Yoldia Sea stage ended with a freshwater phase (Andrén et al., 2002).

The shallowing outlets in the west forced the water level in the Baltic basin to rise and around 10.7 ka BP the next stage, known as the freshwater Ancylus Lake stage, began. For a period of ~500 years, the level of the Ancylus Lake continued to rise to as much as 10 m above sea level but at 10.2 ka BP the lake level fell again (Björck, 1995). This lake level drop occurred because the Ancylus Lake found an outlet in the southern Baltic basin through Mecklenburg Bay and Fehmarn Belt and out through the Great Belt to Kattegat (Fig. 1b) as a complex river system with river channels, levées, and lakes (Björck et al., 2008).

For the following period a couple of hundred years freshwater flowed out through this southern outlet, blocking seawater inflow. However, at 9.8 ka BP a first weak marine influence was recorded in both the Blekinge archipelago (Berglund et al., 2005) and in the Bornholm basin, and, although weaker, also in the Eastern Gotland basin (Andrén et al., 2000a, 2000b). This marks the start of a > 1000 years long transitional phase, the Initial Littorina Sea, characterized by a weak and fluctuating salinity, indicating that the Baltic basin was at level with the sea (Andrén et al., 2000b).

Between 8.5 and 8 ka BP the Öresund Strait (Fig. 1b) was flooded as a result of ongoing global sea level rise, establishing a permanent connection between the Baltic Sea and the North Sea through the Danish straits. This created the modern brackish/marine Baltic Sea, which is also referred to as the Littorina Sea (e.g. Andrén et al., 2011). This transition from a freshwater lake to the modern brackish/marine Littorina Sea is generally referred to as the Ancylus-Littorina transition (A/L transition; e.g. Sohlenius et al., 2001; Jilbert and Slomp, 2013). Since the onset of the Littorina Sea, ongoing glacio-isostatic rebound

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