The East Gotland Basin (Baltic Sea) as a candidate Global Boundary Stratotype Section and Point for the Anthropocene series

Abstract

The short sediment core EMB201/7-4 retrieved from the East Gotland Basin, central Baltic Sea, is explored here as a candidate to host the stratigraphical basis for the Anthropocene series and its equivalent Anthropocene epoch, still to be formalized in the Geological Time Scale. The core has been accurately dated back to 1840 CE using a well-established event stratigraphy approach. A pronounced and significant change occurs at 26.5 cm (dated 1956 ± 4 CE) for a range of geochemical markers including $^{239+240}$Pu, ^{241}Am, fly-ash particles, DDT (organochlorine insecticide), total organic carbon, and bulk organic carbon stable isotopes. This stratigraphic level, which corresponds to a change in both lithology and sediment colour related to early anthropogenic-triggered eutrophication of the central Baltic Sea, is proposed as a Global Boundary Stratotype Section and Point for the Anthropocene series.

Key words: anoxic marine basin, biomarkers, eutrophication, GSSP, microplastics, organic-rich sediments, radionuclides, radiocarbon, SCPs

Introduction

Semi-enclosed basins such as the Baltic Sea are highly impacted by anthropogenic pressures in their catchment areas. Well-known consequences of human-driven eutrophication impacts on the Baltic Sea ecosystem are water deoxygenation and the spread of hypoxic or “dead” zones in deep basins and coastal areas (Gustafsson et al., 2012; Carstensen et al., 2014; Andersen et al., 2017; Reusch et al., 2018). Eutrophication is defined as the enrichment of water by nutrients (nitrogen, phosphorus), which causes an accelerated growth of algae and plants that produce an undesirable disturbance in the balance of organisms present in the water and to the quality of the water concerned (OSPAR, 1998). It reduces the amount of oxygen in water due to the decomposition of sinking organic material by oxygen consuming bacteria, and can eventually lead to benthic ecosystem collapses.

Because of eutrophication, the East Gotland Basin (EGB; 249 m water depth at its deepest point), located in the central Baltic Sea, has been almost permanently hypoxic below 80 m (i.e. water oxygen concentration <2 ml l^{-1}) since the 1950s, and even euxinic in its deepest part (Gustafsson et al., 2012; Carstensen et al., 2014). The expansion of oxygen deficient bottom waters in the central Baltic Sea is interrupted for a few months only during Major Baltic Inflows (MBIs), which are intrusions of oxygen-rich waters from the North Sea (Dellwig et al., 2018; Mohrholz, 2018). MBIs are occasional and occur only when long-lasting easterly winds causing a below-normal Baltic sea level are followed by strong westerly winds (Mohrholz et al., 2015). These inflow events are recorded in sediments through the formation of Mn carbonate (or Ca-rich rhodochrosite) and/or through significant increase in specific trace metals such as Co and V (Dellwig et al., 2019; Dellwig et al., 2021). Except for these mm-thick Mn carbonate-rich layers, a hemipelagic-type of sedimentation with a high content of organic carbon (5 – 18%) and relatively high sedimentation rates (0.3 – 0.5 cm yr^{-1}) has prevailed in the EGB since 1950 CE (Struck et al., 2001; Ilus et al., 2007; Kaiser et al., 2020; Lin et al., 2021). The mean sinking velocity of particles (diatoms) has been estimated around 15 to 70 m d^{-1} (Passow, 1991), which means a particle at the sea surface needs 3 to 16 days to reach the seabed of the EGB. Anoxic conditions in bottom waters of the EGB prevent benthic colonization and bioturbation, and enable the formation of mm-scale laminated sediments in an undisturbed stratigraphic sequence after 1950 CE (Ilus et al., 2007). In contrast to annually or seasonally laminated lake sediments, multiple laminations in sediments from the central Baltic Sea are not annual but controlled by several factors so the composition of single laminae also varies (Sohlenius et al., 1996; Lepland and Stevens, 1998; Brenner, 2001). Before the mid-20th century, the sediment is grey, homogeneous and silty-clayey. Recent sediments (<180yr) from the EGB are thus well-suited to record precisely and continuously the natural and anthropogenic development of the Baltic Sea and its catchment area. Here, we show that the Baltic Sea represents a high quality candidate to host the onset of the Anthropocene via selection of a Global boundary Stratotype Section and Point (GSSP) for

the base of the Anthropocene, that would comprise both a geochronological unit (an Anthropocene epoch) and a chronostratigraphic unit (an Anthropocene series) (Head et al., 2021). The preparatory activities of the Anthropocene Working Group, including events leading to the submission of GSSP proposals and the binding decision that the base of the Anthropocene should align with stratigraphic signals dating to the mid-20th century, are detailed in the introductory article to this special issue (Waters et al., in submission).

Materials and methods

Geographic settings

The Baltic Sea, the world second largest brackish sea, is a shallow, marginal sea with a mean depth of 55 m, and is characterized by an estuarine-like circulation (Elken and Matthäus, 2008). The upper water layer (0 – 60 m) has low salinities (7-8 g kg^{-1}) caused by high freshwater inputs by rivers in the eastern and northern parts of the drainage basin. As a counterpart, relatively high saline water from the North Sea flows in through the Danish Straits. This dense water spreads into deep layers and causes a strong vertical salinity gradient in the central Baltic Sea, with a pronounced halocline around 60–80 m depth. The inflow of saline waters is balanced by upwelling, diapycnal mixing and the outflow of brackish water in surface layers. The catchment area of the Baltic Sea covers 1.7 million km^2, that is more than four times the area of the Baltic Sea itself (369,000 km^2), and is drained by > 200 rivers (Håkanson et al., 2003; Snoeijs-Leijonmalm and Andrén, 2017). About 85 million people live within the Baltic Sea catchment area, and 18% of them within 10 km of the coast (HELCOM, 2010). Agriculture land areas are concentrated mainly in the southern part of the drainage basin, where they occupy >65% of the land area (Snoeijs-Leijonmalm and Andrén, 2017).

Field collection, core scanning, and sampling

Sediment core EMB201/7-4 (57°16.980'N, 020°07.228'E; 241 m water depth; Figure 1) was recovered from the EGB on December 6, 2018, during expedition EMB201 on board the research vessel Elisabeth

Mann Borgese. The coring site was located in the exclusive economic zone of Latvia. A multicorer, equipped with 60 cm-long polyvinyl chloride tubes, was used as this coring device keeps the water-sediment interface undisturbed. The 45 cm-long core was opened lengthwise on board and kept refrigerated (4°C) in the core repository of the Leibniz Institute for Baltic Sea Research (IOW, Germany). Both core halves were scanned at the IOW with an ITRAX X-ray fluorescence (XRF) scanner (Cox Analytical Systems) equipped with a Cr tube operated at 30 kV and 55mA (exposure time of 5 s) with a 200 µm resolution (Table S1). X-ray radiography was recorded at 60 kV and 30 mA, with an exposure time of 225 milliseconds, and at a 200 µm resolution, to identify variations in density along the sediment core. In June 2020, one half of the sediment core was sampled at the IOW in an enclosed laminar flow workstation to prevent airborne contamination. Due to the high water content of recent sediments in the upper part of the core, and to guarantee sufficient amount of sediments for all analyses, the sampling strategy included slicing the core every 1 cm from 0 to 27 cm depth, and every 0.5 cm from 27 to 45 cm depth. This resulted in a total of 63 samples, which were freeze-dried and homogenized before analysis. The second half of the core is kept in the IOW repository as an archive.

Chronological control

An event stratigraphy approach has been developed to accurately date Baltic Sea recent sediments (Dellwig et al., 2018; Kaiser et al., 2020; Dellwig et al., 2021; Lin et al., 2021). This method was applied with modifications in the present study to establish the age model of core EMB201/7-4. Artificial radionuclides ($^{239+240}$Pu, ^{241}Am, ^{137}Cs) here were not used as time markers, but as anthropocene proxies, except ^{137}Cs for the Chernobyl accident in 1986 (see below).

Anthropocene proxies

Relevant proxies used to define the onset of the Anthropocene epoch follow recommendations from Zalasiewicz et al. (2019).

^{241}Am, ^{137}Cs, and ^{210}Pb. Artificial radionuclides ^{210}Am, ^{137}Cs, and natural ^{210}Pb were analysed at the IOW, Germany, by gamma spectrometry with a Ge-well detector (GCW4021-7500SL-RDC-6-ULB). Raw data were then processed with the GENIE 2000 software (Canberra Industries Inc., USA). Counting statistics were better than 5 – 10% for ^{210}Pb and ^{137}Cs and better than 20% for ^{241}Am activities, respectively. The radionuclide activities were calculated using the following standard reference materials (decay corrected): IAEA-447 (^{137}Cs, ^{210}Pb, ^{226}Ra), IAEA-384 (^{241}Am) and IAEA-385 (^{241}Am, ^{137}Cs, ^{226}Ra). The energies used for quantification of isotopes were 46.5 keV for ^{210}Pb, 295 keV and 351 keV for ^{226}Ra, 59.5 keV for ^{241}Am, and 661 keV for ^{137}Cs (Moros et al., 2017). A total of 49 samples was analysed, that is all samples between 0 and 31.5 cm and every second sample between 32 and 44.5 cm (Table S2).

^{238}Pu, ^{239}Pu, and ^{240}Pu. Pu isotopes were analysed at the GAU-Radioanalytical Laboratories of the University of Southampton, UK. Briefly, sediment samples (n = 20; Table S2) were transferred to glass beakers and ignited at 450°C to remove organic matter. The remaining inorganic fraction was spiked with ^{242}Pu chemical recovery tracer, mixed with lithium metaborate flux and fused at 1100°C to achieve a homogeneous melt. The obtained liquid glass was then quenched in deionised water and the resulting solidified glass was dissolved in 8M HNO_3 at 90°C (overnight). The resulting solution was cooled to room temperature and the precipitated boric acid, resulting from the added flux, was filtered off using a GF/A filter paper. Pu was pre-concentrated from the sample solution by means of co-precipitation with $Fe(OH)_3$ at pH 5-6. The precipitate was separated by centrifuging and re-dissolved in 9M HCl. These solutions were transferred to anion exchange columns, previously pre-conditioned with 9M HCl. Pu was retained on the columns and was purified by sequential washes with 9M HCl, 8M HNO_3 and again with 9M HCl. Finally, the Pu fraction was eluted to pre-cleaned beakers using 0.1M NH_4I/9M HCl solution. The solution was evaporated to dryness with HNO_3 added to remove excess iodine present and thin alpha spectrometric sources were prepared by electrodeposition. Each source was then counted using Octete (Ortec/Amtek) alpha spectrometers equipped with passivated implanted planar silicon (PIPS) detectors.

The resulting spectra were analysed using Maestro spectral analysis software. Following counting, $^{240}Pu/^{239}Pu$ ratios were determined using a Thermo Scientific Neptune Plus MC-ICP-MS after radiochemical re-purification of the Pu alpha spectrometry discs (Łokas et al., 2022).

Radiocarbon (^{14}C). ^{14}C was analysed by accelerator mass spectrometry (AMS) at the ETH Zürich, Switzerland. Sediment samples (n = 20; Table S2) were selected between 1 and 31 cm depth, where the highest ^{14}C activity was expected. Each sample (0.1 – 0.3 g) was subject to acid-base-acid treatment (Hajdas, 2008; Hajdas et al., 2021). Briefly, in the first acid step bulk sediment was decarbonated with 0.5M HCl to remove inorganic carbon. After a washing to neutral with MilliQ water the decarbonated sediment was treated with 0.1M NaOH. The liquid was extracted and a few ml of 0.5 M HCl were added to precipitate the base-extracted organic fraction (or humic acid fraction), and freeze dried. The residue, i.e., the base-insoluble organic fraction (or humin fraction), was washed to neutral, treated with 0.5M HCl, washed again to neutral, and freeze-dried. Both fractions were weighed into Al cups for combustion in an elemental analyser (Vario Micro Cube; Elementar) and subsequent graphitization (Nemec et al., 2010). The graphite powder was pressed into the Al cathodes to measure $^{14}C/^{12}C$ and $^{13}C/^{12}C$ ratios using the AMS system of MICADAS (Synal et al., 2000; Wacker et al., 2010). Results are reported as fraction modern $F^{14}C$ (Reimer et al., 2004) and radiocarbon ages (Before Present, BP, where 0 BP = 1950 CE; (Stuiver and Polach, 1977). For an overview of the method and data reporting see Hajdas et al. (2021).

Fly-ash. Spheroidal carbonaceous fly-ash particles (SCPs) were analysed at University College London, UK (Rose, 1994). Briefly, sediments (0.2 g; n = 52; Table S2) were subjected to sequential chemical attack by mineral acids to remove unwanted fractions leaving a suspension of mainly carbonaceous material in water. A known fraction of this final suspension was evaporated onto a coverslip and mounted onto a microscope slide. SCPs were then counted using a light microscope at x450 magnification and the sediment content calculated in units of SCPs per gram of sediment dry weight (SCPs g^{-1} dw). The criteria for SCP identification under the light microscope followed (Rose, 2008). The detection limit for the

technique is typically ca. 100 SCPs g^{-1} dw and accuracy is ca. ± 45 SCPs g^{-1} dw (Rose, 2008) although detection limits were slightly higher in this core (ca. 300 SCPs g^{-1} dw). SCPs were sub-divided into size classes during counting. Analytical blanks and SCP reference material were included in duplicate and triplicate, respectively. Reference materials agreed closely with expected values (6005 ± 70 SCPs g^{-1} dw), while no SCPs were observed in blanks. SCPs were not analysed between 2 and 9 cm due to the low amount of available sediment.

Microplastics. Microplastics (MPs) sampling strategy aimed to maximize MP particles recovery by merging consecutive layers in the core to reach a minimum of ca. 1 g sediment dw, resulting in a total of 20 samples (Table S2). Potential MPs (particle size between 11µm and 5mm) were extracted and isolated from sediments at the IOW, Germany, following a well-stablished protocol (Enders et al., 2020). Briefly, sediments were submitted to a density separation by using sodium polytungstate (1800 kg m^3), which allows all plastic particles to float, and digested with 30% H_2O_2 to eliminate organic particles. The method recovery rate has been estimated to 80%. Automated particle-based analysis was performed at the Biological Institute Helgoland (BAH) of the Alfred Wegner Institute, Helmholtz Centre for Polar and Marine Research, Germany, following a well-established protocol (Primpke et al., 2018). Purified samples were submitted to a second density separation and digestion steps (Abel et al., 2021) to disaggregate flocculates and eliminate potential biofilm immediately before micro-Fourier transform infrared spectroscopic analysis (µFTIR; Hyperion 3000m, Bunker Optics GmbH). This technique measured 100% of remaining particles in each sample to ensure all potential MP particles could be associated to a specific polymer type, or excluded from being classified as plastic. Both extraction and spectroscopic analysis followed protocols to prevent contamination during all steps, including processing blanks to allow estimation of false-positive MP particles that were excluded from final results after interpretation. Contamination from the core tube was not observed.

Persistent organic pollutants and biomarkers. Polycyclic aromatic hydrocarbons (PAHs), dichlorodiphenyltrichloroethane (DDT) and biomarkers (sterols) were analysed at the IOW, Germany (Kanwischer et al., 2020; Kaiser and Lerch, 2022). Briefly, homogenized sediments (0.3 – 0.8 g dw; n = 62; Table S2) were extracted with a DCM:MeOH mixture using an Accelerated Solvent Extraction device (Dionex™ASE™ 350; Thermo Fisher Scientific). After the addition of internal standards, the extracts were desulfurized with activated copper and separated into four fractions by microscale flash column chromatography using hexane (F1), hexane:DCM (F2), DCM (F3) and DCM:MeOH (F4) as eluting solvents (Kaiser and Arz, 2016). The F1 and derivatized F4 fractions were analysed by gas chromatography - mass spectrometry (GC-MS; Agilent Technologies 7890 B GC system and 5977 B Mass Selective Detector). PAHs, p,p'-DDD (1,1,-dichloro-2,2,-bis(p-chlorophenyl)ethane) and p,p'-DDE (1,1-dichloro-2,2-bis(chlorophenyl)ethylene) were identified by comparing retention time with certified external standards (DRE-L20950009CY and DRE-L18000014CY). PAHs and DDT, considered here as the sum of its two degradation products, p,p'-DDD and p,p'-DDE, were quantified using calibration curves based on six calibrator points and expressed as ΣPAH and ΣDDT. Brassicasterol (24-methylcholesta-5,22E-dien-3β-ol), tetrahymanol (gammaceran-3β-ol), coprostanol (5β-cholestan-3β-ol), epi-coprostanol, 5β-stigmastanol (24β-ethyl-5β-cholestan-3β-ol), and epi-5β-stigmastanol were identified by chromatographic and mass spectral comparison with published data. The analytical recovery was estimated to 90% and the limit of quantification to 5 ng g^{-1} sediment dw. Results were normalized to the total organic carbon content for each sample (µg $gTOC^{-1}$).

Total carbon (TC), total organic carbon (TOC), total nitrogen (TN), and C/N ratio. Bulk sediment TC, TOC and TN were measured at the IOW, Germany (Müller and Voss, 1999; Voss et al., 2000). The samples (30 mg dw; n = 62; Table S2) were analysed with a Carlo Erba Flash EA elemental analyser. TOC and TN were measured by weighing the sediment into Ag boats and treating it with 2N HCL until all carbonate compounds had reacted out. The sample was dried at 60°C, pelletized and measured. The

calibration material for C and N analysis was acetanilide (Merck). The analytical precision was better than 1% for TC and TOC, and better than 4% for TN. The C/N ratio was defined as the molar ratio between TOC and TN: C/N = (TOC/12) / (TN/14). Data were not corrected for salt content due to low salinity of the Baltic Sea (Snoeijs-Leijonmalm and Andrén, 2017).

Stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopes. Bulk $\delta^{15}N$ and $\delta^{13}C$ were analysed at the IOW, Germany. Briefly, untreated and treated (acidified) fractions of sediments (n = 62; Table S2) were analysed following Müller and Voss (1999) and Voss et al. (2000). The isotopic compositions were determined using flash combustion in a EA Isolink CN (Thermo Fisher Scientific) elemental analyser at 1020°C coupled via a ConFlo IV (Thermo Fisher Scientific) split interface to a Delta$^+$ V advantage (Thermo Fisher Scientific) isotope ratio mass spectrometer. The isotopic values are reported relative to atmospheric N_2 ($\delta^{15}N$) and Vienna PeeDee Belemnite (VPDB; $\delta^{13}C$). The reference materials used for stable isotope analysis were IAEA-N1, IAEA-N2, IAEA-N3, NBS 22, IAEA-CH-3 and IAEA-CH-6. The analytical precision for both stable isotope ratios was <0.2‰. To correct for the Suess effect, that is a change in $\delta^{13}C$ by the admixture of large amounts of fossil-fuel derived CO_2, the following equation (Schelske and Hodell, 1995; Verburg, 2006) was applied:

$$\delta^{13}C_{org.corr} = \delta^{13}C_{org} - (4{,}577.8 - 7.3430 * y + 3.9213 \times 10^{-3} * y^2 - 6.9812 * 10^{-7} * y^3)$$

where y = year CE.

Inorganic geochemical markers. Sedimentary major and trace element contents and stable Pb isotopes were analysed at the IOW, Germany. Briefly, the sediments (100 mg dw; n = 62; Table S2) were digested with a HNO_3-$HClO_4$-HF mixture in closed Teflon vessels at 180°C for 12 h and the residues were fumed-off 3-times with 6 M HCl and finally diluted with 2 vol% HNO_3 (Dellwig et al., 2019). The content of Al was measured by inductively coupled plasma optical emission spectrometry (ICP-OES, iCAP 7400 Duo, Thermo Fisher Scientific) using external calibration and Sc as internal standard. Analytical precision and accuracy were estimated with the international reference material SGR-1b (USGS) and were better than

3.8% and -3.9%, respectively. Trace metals Cu, Pb (including ^{206}Pb and ^{207}Pb) and Zn were determined by inductively coupled plasma mass spectrometry (ICP-MS, iCAP Q, Thermo Fisher Scientific) coupled to a prep*FAST* system (Elemental Scientific) using He as collision gas, external calibration, and Rh and Ir as internal standards. Analytical precision and accuracy of concentration measurements were also checked with SGR-1b and were better than 3.3% and -3.4%, respectively. For stable Pb isotope ratios, NIST SRM-981 was used resulting in a precision and accuracy of 0.31% and -0.04%, respectively (Dellwig et al., 2021). Results were normalized to Al content for each sample to minimize dilution effects caused by organic matter, carbonate and/or opal. To determine Hg contents, sediments (100 mg dw; n = 63; Table S2) were analysed with a Milestone DMA-80 EVO Direct Mercury Analyser by thermal decomposition at 750 °C, gold amalgamation and atomic absorption spectrophotometry at 253.65 nm. Data were calibrated against the BCR 142R certified reference material and a Baltic Sea reference sediment. Analytical precision based on reference materials was better than 6%.

Numerical analysis. To detect positions of abrupt change (changepoints) within the different datasets, a modelling approach (Gallagher et al., 2011) was applied using the software Past v4.10 (Hammer et al., 2001). Before analysis, the datasets were interpolated every 1 cm, missing values were treated by linear interpolation, and mean and standard deviation were automatically normalized in order to equally weigh each dataset. The default value of 100,000 iterations (Markov chain Monte Carlo) was used and the number of changepoint set to n = 1 (Table S3).

Results

Lithology

The lithology of core EMB201/7-4 was characterized by a pronounced visual transition at around 27 cm sediment depth (Figure 2). Between 45 – 27 cm depth the sediment was grey, relatively homogeneous, silty-clay, with low organic carbon and water contents. Between 27 – 0 cm depth it was brown to dark brown and black, cm-thick layered and sometimes laminated, with high organic carbon and water

239 contents. Note that, after oxidation, sediment colours change to dark grey below 27 cm and to brown to
240 dark brown between 27 – 0 cm. The upper part of the core was also characterized by a few light grey,
241 mm-wide, clayey to silty laminations mainly between 21 – 25 cm depth.
242 *Chronology*
243 The use of unsupported ^{210}Pb ($^{210}Pb_{unsupp}$) to date recent sediments is challenging in the Baltic Sea because
244 the depositional environment and the sedimentation type have varied over the past 100 years, resulting in
245 an irregular decrease in $^{210}Pb_{unsupp}$ (Mattila et al., 2006; Moros et al., 2017; Häusler et al., 2018). Seven
246 parameters were considered to build the stratigraphy ($^{206/207}$Pb, Hg, ΣPAH, ΣDDT, Mn/Ti, Co/Al and
247 ^{137}Cs; Figure 2b-h), and twelve stratigraphical events, or time markers, were used to date the sediment
248 (Table 1). The uncertainties of the time markers were estimated following Moros et al. (2017), Kaiser et
249 al. (2018, 2020), and Lin et al. (2021). The early increases in $^{206/207}$Pb, Hg and ΣPAH reflect the early
250 increase in wood and coal combustion around 1870 CE related to the beginning of the Second Industrial
251 Revolution, or Technological Revolution (Hites Ronald et al., 1977; Elmquist et al., 2007; Díaz-Somoano
252 et al. 2009; Hanke et al., 2019). The abrupt rise in ΣDDT is likely related to increased DDT use for
253 agriculture since 1950 CE (Li and Macdonald, 2005), a well-known pattern in Baltic Sea and European
254 lake sediments (Thevenon et al., 2013; Sabatier et al., 2014). Mn and Co enrichments are attributed to
255 four MBIs (1978, 1994, 2003 and 2014 CE (Dellwig et al., 2021; Lin et al., 2021), and tentatively to other
256 four MBIs (1964, 1967, 1970 and 1972 CE; Figure S1). The ^{137}Cs increase has been attributed to the
257 Chernobyl accident in 1986 CE. The year of the core recovery (2018 CE) was attributed to the core top
258 layer. Linear sedimentation rates were assumed between time markers resulting in a mean sedimentation
259 rate increasing from 0.2 cm yr^{-1} between 45 cm (ascribed the date 1840 ± 10 CE) and 28.4 cm depth
260 (1950 ± 4 CE) to 0.4 cm yr^{-1} from 28.4 cm to the top of the core (Figure 2g).
261 *Radioisotopes (actinides and ^{14}C)*

^{241}Am and $^{239+240}$Pu profiles showed similar patterns (Figure 3a-b). Both radionuclides were close to the detection limit below 28.8 cm depth (1947 ± 4 CE). They started to increase at 27.3 cm depth (1953 ± 4 CE) and reached a maximum at 24.5 cm depth (1963 ± 4 CE) with activities of 8.4 Bq kg^{-1} for ^{241}Am and 0.025 Bq kg^{-1} for $^{239+240}$Pu. Above this depth, the activities of both radionuclides decreased abruptly. A second, sharp peak occurred at 15.5 cm depth (1986 ± 2 CE) in both radionuclide profiles. Values of $^{240/239}$Pu (Figure 3c), a tracer for Pu source, were >0.301 at 28.8 and 28.3 cm depth (1947 ± 4 and 1950 ± 4 CE, respectively) and between 0.160 – 0.219 from 27.8 cm depth (1952 ± 4 CE) except for slightly higher values at 15.5 cm (1986 ± 2 CE) and 10.5 cm depth (1998 ± 2 CE). F^{14}C (Figure 3d), the normalized fraction of ^{14}C (Hajdas et al., 2021), was relatively stable (~ 0.8) between 30.8 and 27.3 cm depth (1933 – 1953 CE). It started increasing at 26.5 cm depth (1956 ± 4 CE), reached a maximum of 1.2 between 23.5 and 20.5 cm (1966 – 1974 CE), and decreased to 1.0 at 1.5 cm depth (2014 ± 1 CE).

Spheroidal carbonaceous particles. A first appearance of SCPs (Figure 4a) occurred at 36.3 cm depth (1897 ± 8 CE), but SCP contents remained low and sporadic until 27.3 cm depth (1953 ± 4 CE). Only SCPs <25 µm (and frequently <10 µm) were observed in this lower section. Above 27.3 cm depth, SCP contents increased, initially rapidly, through to a peak of >7,700 SCPs g^{-1} dw at 16.5 cm depth (1983 ± 3 CE). From this peak, SCP contents showed a general decline through to the top of the core. Above 28.3 cm, larger SCPs appeared for the first time and SCPs had a wide size range from <5 µm to >50µm (Table S2).

Microplastics. A total of 178 MP particles and six different polymers were identified. Most MP particles were smaller than 100 µm in size, and fibres were almost absent. MP amounts (Figure 4b) ranged between 248 and 7,135 MPs kg^{-1} dw. MPs were identified at all depths between 42.3 and 6.5 cm (1857 – 2005 CE), with a maximum abundance at 20.5 cm depth (1974 ± 4 CE). In terms of polymer diversity, a near-consistent pattern was observed within all layers with predominance of ethylene-propylene-diene monomer rubbers (Table S2).

Polycyclic aromatic hydrocarbons and dichlorodiphenyltrichloroethane. The most abundant PAHs were phenanthrene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[ah]anthracene and benzo[ghi]perylene. Their sum (ΣPAH) increased gradually from 41.3 to 17.5 cm depth (1863 – 1981 CE), reaching a maximum around 170 µg $gTOC^{-1}$, and decreased gradually to reach a minimum of 20 µg $gTOC^{-1}$ near the top of the core (Figure 4c). ΣDDT (Figure 4d) was below detection limit between 44.8 and 28.8 cm depth (1840 – 1947 CE). It increased rapidly at 28.3 cm depth (1950 ± 4 CE) to reach a maximum of 20 µg $gTOC^{-1}$ between 25.5 and 21.5 cm depth (1960 – 1971 CE). After the peak, ΣDDT decreased gradually towards the top of the core to reach contents <1 µg $gTOC^{-1}$.

Total carbon, total organic carbon, total nitrogen, total organic nitrogen, and the C/N ratio. Both TC and TOC values (Figure 5a-b) were relatively constant around 3-4% between 44.8 and 29.8 cm depth (1840 – 1940 CE). Above 29.8 cm depth (1940 ± 5 CE), they increased gradually to reach 19.8 and 17.3% at the top of the core, respectively. TC and TOC showed a significant positive linear correlation ($r^2 = 0.97$). TN values were <0.5% below 27.8 cm depth (1952 ± 4 CE), followed by a gradual increase to the top of the core, where the values reached 2% (Figure 5c). The C/N ratio (Figure 5d) ranged between 10 and 12 below 27.8 cm depth (1952 ± 4 CE), and between 8 – 10 above 23.5 cm depth (1966 ± 4 CE).

Stable carbon and nitrogen isotopes. The record of bulk organic matter $\delta^{13}C$ ($\delta^{13}C_{org}$) presented a -1.2‰ shift from -26.2 ± 0.2‰ below 27.8 cm depth (1952 ± 4 CE) to -25.0 ± 0.3‰ above this depth (Figure 5e). Both $\delta^{13}C_{org}$ and $\delta^{13}C_{org.corr}$ vertical profiles (Figure 5e) were relatively similar, except above 11.5 cm depth (1996 ± 2 CE), where $\delta^{13}C_{org}$ values decreased gradually compared to $\delta^{13}C_{org.corr}$. $\delta^{15}N$ values presented a relatively large scatter (Figure 5f). A general increasing trend of +2‰ occurred between 44.8 – 27.8 cm depth (1840 – 1952 CE). Above 26.5 cm depth (1956 ± 4 CE), $\delta^{15}N$ values fluctuated around 3.3 ± 0.4‰.

309 *Inorganic geochemical signals.* Hg/Al, Cu/Al, Zn/Al, and Pb/Al values (Figure 6a-d) increased from 28.8
310 cm depth (1947 ± 4 CE). The ratios were highest between 24 – 18 cm depth (1966 – 1978 CE), and
311 decreased until 16.5 cm depth (1983 ± CE). Cu/Al and Zn/Al increased again above 16.5 cm depth (1983
312 ± 3 CE). $^{206/207}$Pb values (Figure 6e) decreased gradually from 1.253 to 1.164 between 38.8 – 22.5 cm
313 depth (1873 – 1969 CE) with a slight increase from 33.3 to 31.8 cm depth (1917 – 1927 CE). Above 22.5
314 cm depth (1969 ± 4 CE), $^{206/207}$Pb values increased and reached a value of 1.209 at the top of the core.
315 *Biotic markers.* Brassicasterol, contents increased significantly from 1.5 to 8.5 µg $gTOC^{-1}$ between 26.5
316 cm depth (1956 ± 4 CE) and the top of the core (Figure 7a). Tetrahymanol contents (Figure 7b) showed a
317 similar trend with very low values below 26.5 cm depth (1956 ± 4 CE) followed by an increase until 10.5
318 cm depth (1998 ± 2 CE). Above this depth, the contents were relatively stable until the top of the core.
319 Faecal lipids (the sum of coprostanol and epi-coprostanol) first appeared at 26.5 cm depth (1956 ± 4 CE)
320 and increased until the top of the core (Figure 7c). The R2 ratio (R2 = coprostanol + epicoprostanol / 5β-
321 stigmastanol + epi-5β-stigmastanol + coprostanol + epicoprostanol; Kaiser and Lerch 2022) ranged
322 between 0.4 – 0.8 and increased from 26.5 cm (1956 ± 4 CE) to the top of the core (Figure 7d; Table S2).
323 **Discussion**
324 *Chronology.* The age model of core EMB201/7-4 (Figure 2) is well-constrained by eleven stratigraphic
325 events between 1950 – 2018 CE (28.3 – 0 cm depth). Most of these stratigraphic events are known in the
326 Baltic Sea and have already been used to date recent sediments (Moros et al., 2017; Häusler et al., 2018;
327 Kaiser et al., 2020; Dellwig et al., 2021; Lin et al., 2021). Before 1950 ± 4 CE (28.3 cm depth), however,
328 the age model is based on a single stratigraphic event at 1870 ± 10 CE (40.3 cm depth) and on a linear
329 extrapolation to the bottom of the core dated 1840 ± 10 CE (45 cm depth). Between 1870 – 1950 CE,
330 older-than-expected ^{14}C ages (1892 ± 80 year BP; n = 7) were obtained (Table S2). This indicates an
331 admixture of relatively older organic matter most likely due to the inwash of pre-aged terrestrial organic
332 matter from the drainage basin (Howarth et al., 2013; Schroeter et al., 2021), or sediment reworking

(Moros et al., 2020). The $^{206/207}$Pb and ΣPAH profiles from core EMB201/7-4 (Figure 2b,d) support the absence of sediment reworking as they are comparable to existing records from lakes within the Baltic Sea drainage basin (Renberg et al., 2001, 2002). Therefore, the inwash of pre-aged terrestrial organic matter likely explains older-than-expected radiocarbon ages, but the sediments were not affected significantly by reworking before 1950 CE. The event stratigraphy approach followed here represents the most advanced method to date Baltic Sea recent sediments.

Radionuclides as primary markers for the Anthropocene GSSP level in Baltic Sea sediments. For the Anthropocene to be a formal chronostratigraphic/geochronologic unit of geological time, a synchronous base is needed (Head et al., 2021; Waters et al., in submission). $^{239+240}$Pu and ^{241}Am artificial radionuclides produced by atmospheric nuclear bomb tests are the main candidates as primary markers in the context of the Anthropocene because of their longevity (half-lives of 24,110 and 432 years, respectively) and appearance around 1950 CE in different archives worldwide (Zalasiewicz et al., 2017; Waters et al., 2018; Waters et al., 2019). While $^{239+240}$Pu was first identified above detection limit in 1947 ± 4 CE (28.8 cm depth) in core EMB201/7-4, both ^{241}Am and $^{239+240}$Pu (Figure 8a-b) started to increase markedly after 1953 ± 4 CE (27.3 cm depth). They reached maximum values in 1963 ± 4 CE (24.5 cm depth), when the Limited Test-Ban Treaty was established, which was followed by a rapid decline in radionuclide fallout worldwide (Waters et al., 2018; Waters et al., 2019). $^{240/239}$Pu values (Figure 3c) above 0.30 between 1947 – 1950 CE (28.8 – 28.3 cm depth) suggested a main radionuclide source from test sites in the Pacific Ocean (Pacific Proving Grounds; (Koide et al., 1985; Buesseler, 1997; Muramatsu et al., 2000; Lachner et al., 2010), while values below 0.19 since 1953 ± 4 CE (27.3 cm depth) are typical of global fallout (Buesseler and Sholkovitz, 1987; Kelley et al., 1999). Slightly increased values in 1986 ± 2 CE (15.5 cm depth) are consistent with the Chernobyl accident (Muramatsu et al., 2000).

The ^{137}Cs artificial radionuclide (Figure 2h) activity increased slightly from 1956 ± 4 CE (26.5 cm depth) due to atmospheric nuclear weapon tests, but the most prominent increase occurred after 1981 ± 3 CE

(17.5 cm depth) because of the Chernobyl accident in 1986 CE. As documented, the radioactive cloud from this accident first travelled northward and resulted in a high deposition of ^{137}Cs radionuclides in the Baltic Sea region (Mattila et al., 2006; Ilus et al., 2007; Moros et al., 2017; Dellwig et al., 2018). F^{14}C, the atmospheric radiocarbon excess released by nuclear bomb tests, started to increase in 1956 ± 4 CE (26.5 cm depth) and reached maximum values in 1966 ± 4 CE (23.5 cm depth), about three years after ^{241}Am and $^{239+240}$Pu in both cases (Figure 3d). This delay can be attributed to atmosphere-ocean ^{14}C equilibration time (Waters et al., 2019). Also, post-depositional, vertical migration within sediments, sediment remobilization, as well as sediment inwash from the drainage basin are typical potential issues affecting radionuclide profiles (Davis et al., 1984; He et al., 1996; Owens and Walling, 1996; Mattila et al., 2006; Ilus et al., 2007; Hancock et al., 2014; Waters et al., 2019; Haltia et al., 2021; Schroeter et al., 2021). These issues likely explain the detection of ^{137}Cs since 1917 ± 6 CE (33.3 cm depth) because it is very mobile in sediments compared to ^{241}Am, as well as the detection of radionuclides in the sediments until the present-day.

Artificial $^{239+240}$Pu and ^{241}Am radionuclides from nuclear weapons testing are therefore considered here as the most relevant primary markers in the context of the Holocene-Anthropocene boundary. Results from a changepoint model (Figure 8c; Table S3) indicated a change starting in 1948 ± 4 CE (28.5 cm depth), but the main change occurred in 1956 ± 4 CE (26.5 cm depth), which would be the proposed GSSP level in Baltic Sea sediments.

Anthropogenic pollutants as secondary markers for the Anthropocene GSSP level in Baltic Sea sediments. SCP contents (Figure 8d) increased markedly after 1953 ± 4 CE (27.3 cm depth) due to an increase in coal and fuel oil combustion related to a high electricity demand after WWII. Furthermore, the introduction of new SCP sources is suggested by their wider range of sizes since that time (Table S2). Decreasing SCP contents since 1983 ± 3 CE (16.5 cm depth) reflects the reduction in SCPs release to the atmosphere due to the introduction of particle-arrestor technology since the 1970s and the shift away from

coal and oil fuels to natural gas, nuclear and renewable energy sources (Rose, 2015; Rose and Gałuszka, 2019). The Baltic Sea SCP profile is compatible with other lake sediments reported globally (Rose, 2015).

MP amounts (Figure 4b) increased between 1960 – 1974 CE (25.5 – 20.5 cm depth) as a result of a rapid and intense increase in both production and use of an array of polymer types (Zalasiewicz et al., 2016; Ivar do Sul and Labrenz, 2020). Ethylene-propylene-diene monomer rubbers, the prevalent category within polymers identified here (Table S2), are widely used for a range of marine-and terrestrial-based activities and very common in the marine environment (Primpke et al., 2018; Lorenz et al., 2019). The MP profile suggests that the MP stock in the Baltic Sea marine environment consists of polymers from various provenances affected by different taphonomic processes, and hence does not reflect a direct or straightforward response to world plastic production (Bancone et al., 2020).

Hg, Pb, Cu and Zn are markers of anthropogenic industrial pollution in sediments (Rauch and Pacyna, 2009; Birch et al., 2015; Gałuszka and Rose, 2019). The main trends in Hg/Al, Cu/Al, Zn/Al and Pb/Al (Figure 6a-d) reflect an increase of coal combustion and metal production after the mid-20th century, and a decrease from 1970s CE due to a reduction in industrial dust emission and fossil fuel combustion in northern and western Europe (Rühling and Tyler, 2001; Callender, 2014; Gałuszka and Wagreich, 2019). However, a decrease to background levels in present-day sediments is not observed for most of the metals. This is because trace metals are generally sensitive to changing redox conditions (Bruland et al., 2013), which biases the anthropogenic signal in Baltic Sea sediments. $^{206/207}$Pb is not redox-sensitive, and reflects therefore an unbiased signal (Figure 6e). Decreasing $^{206/207}$Pb values between the early-1870s CE and the late-1960s CE are most likely due to both increasing coal combustion during the Second Industrial Revolution and the spread of leaded gasoline combustion in Europe since 1950 CE (Renberg et al., 2002; Díaz-Somoano et al. 2009; Boyle et al., 2014). Increasing values since the early-1970s (21.5 cm

404 depth) reflect the decline of lead pollution in Europe due to both the implementation of stricter industrial
405 emission standards and the phase-out of alkyl-lead gasoline (Renberg et al., 2001; Renberg et al., 2002).
406 The appearance of the persistent organic pollutant DDT (Figure 8e) in 1950 ± 4 CE (28.3 cm depth) and
407 its pronounced increase up to 1960 ± 4 CE (25.5 cm depth) is related to the increasing use of DDT in
408 Baltic Sea countries after 1945 CE to control insects on agricultural crops. Indeed, the former Soviet
409 Union had the second highest (after the United States) historical DDT usage in agriculture from 1952
410 to1971 CE (Li and Macdonald, 2005). The decrease in DDT content after 1971 ± 4 CE (21.5 cm depth)
411 reflects the ban on its use within Baltic Sea countries since the early-1970s (HELCOM, 2010).
412 Coprostanol and epicoprostanol faecal lipids (Figure 7c-d) first appeared in 1956 ± 4 CE (26.5 cm depth)
413 and increased subsequently as a result of increasing sewage pollution in the central Baltic Sea (Kaiser and
414 Lerch, 2022). The fraction of human-derived faecal lipids was also increasing since 1956 ± 4 CE as
415 suggested by higher R2 values. Therefore, sedimentary faecal lipids reflect the increasing human-derived
416 sewage pollution related to population growth in the Baltic Sea drainage basin (Kaiser and Lerch, 2022).
417 For anthropogenic pollutants, results from the changepoint model (Table S3) indicated a change starting
418 in 1956 ± 4 CE (26.5 cm depth). This change is, however, more pronounced in 1960 ± 4 CE (25.5 cm
419 depth), which corresponds closely to the inflection point of many indicators of the Great Acceleration
420 (Head et al., 2021).
421 *Organic proxies as secondary markers for the Anthropocene GSSP level in Baltic Sea sediments.* The
422 pronounced increase in TOC values (Figure 8f) since 1956 ± 4 CE (26.5 cm depth) reflects the growing
423 burial of phytoplankton biomass under hypoxic conditions due to the eutrophication of the central Baltic
424 Sea deep basins since 1950 CE (Emeis et al., 2000; Struck et al., 2000). Indeed, a shift towards heavier
425 $\delta^{13}C_{org.corr}$ values (Figure 8g) after 1952 ± 4 CE (27.8 cm depth) indicates a higher proportion of
426 phytoplankton biomass, as organisms are forced to use the heavier C isotope (^{13}C) when primary
427 production is high (Mizutani and Wada, 1982; Struck et al., 2000; Teranes and Bernasconi, 2005). A

decrease in C/N values (Figure 5e) after 1960 ± 4 CE (25.5 cm depth) also indicates an increasing fraction of phytoplankton biomass in Baltic Sea sediments (Emeis et al., 2000; Lamb et al., 2006). The contents of brassicasterol (Figure 7a), or "diatomsterol", a biomarker for the occurrence of diatoms, increased substantially after 1952 ±4 CE (27.8 cm depth), also indicating the growth of phytoplankton biomass mainly due to eutrophication. The similar increase in tetrahymanol contents (Figure 7b) reflects the concomitant development of a pelagic redoxcline in the central Baltic Sea as tetrahymanol is a biomarker for ciliates thriving abundantly near the redoxclines of hypoxic environments (Harvey and Mcmanus, 1991; Anderson et al., 2012; Edgcomb and Pachiadaki, 2014; Pachiadaki et al., 2014).

An increasing trend in $\delta^{15}N$ values (Figure 5g) between 1840 – 1950 CE (44.8 – 28.3 cm depth) resulted most likely from the phytoplankton assimilation of land-derived, isotopically heavier nitrate inputs (Struck et al., 2000; Renberg et al., 2001). Indeed, large-scale changes in land use occurred in the Baltic Sea watershed after the beginning of the Second Industrial Revolution (Zillén and Conley, 2010). After 1950 CE, however, $\delta^{15}N$ values started to decrease slightly, possibly due to higher inputs of isotopically lighter nitrogen, and increasing biomass of diazotrophic cyanobacteria, an important contributor to the pelagic N-cycle of the central Baltic Sea since 1950 CE (Voß et al., 1997; Struck et al., 1998; Struck et al., 2000; Kendall et al., 2007; Kaiser et al., 2020). Periods with increasing $\delta^{15}N$ values after 1950 CE may be related to higher inputs of isotopically heavier, wastewater to the Baltic Sea (Kendall et al., 2007).

As shown for anthropogenic pollution markers, the results from the changepoint model (Table S3) suggested that the main change in organic proxies occurred in 1960 ± 4 CE (25.5 cm depth), synchronously with many indicators of the Great Acceleration (Syvitski et al., 2020; Head et al., 2021).

Final remarks regarding the Baltic Sea as GSSP for the Anthropocene series. The present study supports the Anthropocene as a stratigraphically substantiated unit, that should be defined by a GSSP and formalized at the rank of epoch with an inception at around 1950 CE (Zalasiewicz et al., 2017). Artificial radioisotopes released during atmospheric nuclear weapons testing, which are the most significant

markers in the context the Anthropocene, showed a main change in core EMB201/7-4 in 1956 ± 4 CE as indicated by the changepoint model. This point is therefore proposed as the GSSP level for the Anthropocene series (Figure 8). It corresponds to a marked change in lithology and sediment colour from light grey to dark brown (or dark grey to brown after oxidation), which reflects the beginning of eutrophication in the central Baltic Sea due to an increasing anthropogenic pressure. Despite the most accurate approach used to date core EMB201/7-4, the lack of annual lamination may flag the Baltic Sea as a GSSP candidate for the Anthropocene series.

If accepted, the East Gotland Basin GSSP would be in accordance to all 65 GSSPs presently ratified, which occur within strata deposited in marine sedimentary environments with the exception of the three Holocene GSSPs (Walker et al., 2018); Waters et al., 2019). The archived half of core EMB201/7-4 is preserved and accessible at the IOW, Germany. Although marine GSSP sites are not as accessible as terrestrial ones, new sediment cores from exactly the same location can be recovered during expeditions on board research vessels. Finally, if appropriate, the name "Baltician" is suggested as potential stage/age (Luciano, 2022).

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Figure and Table captions

Figure 1. (a) Study area (black rectangle) in the Baltic Sea; the countries, main lakes and rivers of the catchment area - delimited by a bold line - are shown; (b) bathymetric chart (Baltic Sea Bathymetry Database) of the study area with the location of core EMB201/7-4 in the East Gotland Basin (EGB).

Figure 2. Pictures (true colour and radiography), depth profiles of $^{210}Pb_{unsupp}$ (a), $^{206/207}Pb$ (b), Hg (c), ΣPAH (d) and ΣDDT (e) contents, Mn/Ti (f), Co/Al (g) and ^{137}Cs (h), and age-depth relationship (i) of core EMB201/7-4. The assigned time markers are shown by horizontal grey lines and listed in Table 1.

Figure 3. Depth profiles of ^{241}Am (a) and $^{239+240}Pu$ (b) activities, $^{240/239}Pu$ (c), and $F^{14}C$ (d) in core EMB201/7-4.

Figure 4. Depth profiles of SCPs (a), microplastics (b), ΣPAH (c) and ΣDDT (d) contents in core EMB201/7-4.

Figure 5. Depth profiles of TC (a), TOC (b), and TN (c) contents, C/N (d; reversed plotted), $\delta^{13}C_{org}$ and $\delta^{13}C_{org.corr}$ (e), and $\delta^{15}N$ (f) in core EMB201/7-4.

Figure 6. Depth profiles of Hg/Al (a), Cu/Al (b), Zn/Al (c), Pb/Al (d) and $^{206/207}Pb$ (e; reversed plotted) in core EMB201/7-4.

Figure 7. Depth profiles of brassicasterol (a) and tetrahymanol (b) contents, the sum of coprostanol and epicoprostanol contents (c), and the faecal stanol-based R2 ratio (d) in core EMB201/7-4.

Figure 8. Pictures of core EMB201/7-4 (true colour and radiography), primary markers (a-b), and changepoint modelling results (c) used to define the GSSP level in 1956 ± 4 CE (26.5 cm depth; horizontal line). Secondary markers selected to illustrate changes in anthropogenic pollution (d-e) and eutrophication (f-g) in the Baltic Sea are also shown. A close-up (h) indicating the proposed GSSP level at 26.5 cm in the core is represented by a yellow dotted line.

Table 1. List of the time markers used to establish the age model of core EMB201/7-4.

Supplementary data

Table S1. XRF core scanner data (in count per second) of core EMB201/7-4.

Table S2. Datasets of core EMB201/7-4. Note that some datasets were not discussed in the text.

Table S3. Changepoint modelling results.

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Supplemental material

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