1 Title

- 2 The Palmer ice core as a candidate Global boundary Stratotype Section and Point for the
- 3 Anthropocene series.
- 4

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36 Abstract

- 37 The remote Antarctic continent, distant from human industrial activity, should be one of the last
- 38 places on Earth to capture Anthropogenic change. Hence, stratigraphic evidence of pollution and
- 39 nuclear activity in the Antarctic provides proof of the global nature of the Anthropocene epoch. We
- 40 propose an Antarctic Peninsula ice core candidate for the Global boundary Stratotype Section and
- 41 Point (GSSP) to the onset of the Anthropocene. The Palmer ice core captures the first evidence of
- 42 spheroidal carbonaceous fly ash particles (SCPs), resulting from high temperature combustion
- 43 deposited in Antarctic ice. SCPs first appear in 1936 CE, preceding the rise in plutonium (²³⁹⁺²⁴⁰Pu)
- 44 concentrations from 1945 CE onward. GSSP 1952 CE occurs at a depth of 34.9 m, coincident with the
- 45 peak in ²³⁹⁺²⁴⁰Pu the primary marker for this site.

46 Keywords

47 Antarctic Peninsula, Spheroidal carbonaceous particles, ice core, methane, plutonium.

48 Introduction

- 49 Antarctica's remote location, distant from industrial sources and with no permanent or indigenous
- 50 human population, makes it an ideal candidate for identifying the onset of the Anthropocene. Often
- referred to as the "last great wilderness", Antarctica is assumed to be a pristine environment free
- 52 from the influence of human activity. However, the presence of atmospheric pollutants, soot and
- 53 black carbon from fossil fuel and biomass burning (Cordero et al., 2022), heavy metals from mining
- 54 (Potocki et al., 2016; Bargagli, 2008) and microplastics (Aves et al., 2022) are a powerful reminder
- that the Anthropocene is a global phenomenon and even the most remote region on Earth bears its
- 56 mark.
- 57 The Antarctic Peninsula was chosen as a location to identify the Global boundary Stratotype Section
- and Point (GSSP) for the Anthropocene Series due to its potential suitability as a GSSP and sensitivity
- to anthropogenic climate change. The preparatory activities of the Anthropocene Working Group
- 60 (AWG), including events leading to the submission of GSSP proposals and the binding decision that
- 61 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, 2022). In this paper we focus on
 the Palmer ice core from the Antarctic Peninsula, as an invited contribution to the AWG to propose a
- 64 suitable Antarctic ice core GSSP site.
- 65 Here we present the Palmer ice core as a candidate for the Anthropocene series. We provide
- 66 evidence from two Anthropogenic proxies, spheroidal carbonaceous fly ash particles (SCPs) and
- 67 plutonium ($^{239+240}$ Pu). We also include the stable water isotopes (δ^{18} O), snow accumulation and
- 68 atmospheric methane (CH₄) concentrations from this site as supporting material relating to climate
- and anthropogenic change. The Palmer ice core was selected from a transect of ice cores drilled
- along the spine of the Antarctic Peninsula since the mid-1990s (Thomas. and Tetzner., 2018).
- Although arguably many other ice cores could have been proposed to represent the Antarctic GSSP,
- the Palmer ice core was selected based on the following criteria:

73 The exceptional chronology and sample resolution

- 74 The Antarctic Peninsula receives the highest amount of snowfall of all Antarctic regions, exceeding
- 75 ~4 m per year in some areas (van Wessem et al., 2016). Snowfall is highest around the coast and
- areas of orographic uplift, such as the Palmer site. Deposition of anthropogenic proxies is likely
- 77 related to snowfall (wet deposition) and thus the concentration of proxies is expected to be highest

- 78 at coastal and high snow accumulation sites. Evaluation of reanalysis data (ERA-Interim ,1979–2010)
- confirms that the ice core site receives an estimated annual average precipitation minus evaporation
- 80 of 49 cm (water equivalent). While lower elevation Peninsula ice cores have higher snow
- 81 accumulation (Thomas et al., 2017), few extend beyond the past ~150 years (Thomas. and Tetzner.,
- 82 2018; Emanuelsson et al., 2022b). The Palmer ice core was selected to provide sufficient layer
- 83 thickness to capture changes at sub-annual resolution, while ensuring a record that extends over the
- 84 past ~300 years.

85 The sensitivity to Anthropogenic climate change

- 86 In contrast to much of the Antarctic continent, the Antarctic Peninsula is the only region to reveal
- 87 strong and statistically significant warming trends since observations began in 1957 (Jones et al.,
- 88 2016; Gonzalez and Fortuny, 2018). Paleoclimate observations from ice cores and moss peat banks
- suggest that the warming from 1957 onwards is part of a 100-year trend (Thomas et al., 2013; Royles
- 90 et al., 2013) that began in the ~1920s. The rise in surface temperatures is accompanied by significant
- 91 increases in snow accumulation (Thomas et al., 2017; Thomas et al., 2008; Thomas et al., 2015;
- 92 Medley and Thomas, 2019), which has been linked changes in atmospheric circulation, sea ice
- 93 conditions and the increased moisture content associated with a warming atmosphere (Medley and
- 94 Thomas, 2019). The increased surface temperatures, together with the increased occurrence of
- 95 warm and dry fohn winds descending over the Antarctic Peninsula, have been attributed to the
- 96 collapse of Antarctic Peninsula ice shelves (e.g. Banwell et al., 2013).

97 The site has excellent sample preservation, not impacted by melt

- 98 Despite the evidence of increased surface temperatures during the 20th century (e.g. Jones et al.,
- 99 2016), the occurrence of surface melt on the Antarctic Peninsula is limited to the floating ice shelves
- and low-elevation coastal locations (van Wessem at al., 2016). While the James Ross Island ice core,
- 101 from the northern tip of the Antarctic Peninsula, reveals an acceleration in surface melting during
- the 20th century (Abram et al., 2013), there are very few visible melt features in the Palmer core.
- 103 Therefore, the impact of melt on the climate and anthropogenic proxies at Palmer is assumed to be
- 104 negligible. Indeed, a recent study revealed that the stable water isotope record (δ^{18} O) from Palmer
- 105 has the potential to reconstruct melt over the Larsen ice shelves (Emanuelsson et al., 2022a), even
- 106 though the δ^{18} O itself is not directly impacted by melting. Thus, the strategic location of the Palmer
- 107 core makes it sensitive to climate variability, without being adversely impacted by it.

108 The link to long-range transport for anthropogenic proxies

- 109 Back-trajectory analysis confirms that the Amundsen- Bellingshausen Sea is the dominant source
- region for air-masses reaching the Palmer ice core site (Thomas and Bracegirdle, 2015). The
- 111 geographical setting of the Antarctic Peninsula, which forms a barrier for the strong southern
- hemisphere westerly winds, deflects onshore (northerly) air-masses to the Palmer site. An observed
- seasonal migration in the trajectories (1979-2010) is related to changes in the Amundsen Sea Low, a
- 114 climatological low-pressure system that is driven by large-scale modes of climate variability (Thomas
- and Bracegirdle, 2015). These include the Southern Annular Mode (SAM), largest mode of variability
- in the Southern Hemisphere, and El Nino Southern Oscillation (ENSO). The shift to the positive phase
- of the SAM, in response to stratospheric ozone depletion (Lubin et al., 2008), constitutes one of the
- 118 largest trends in the past 200 years (Jones et al., 2016). Antarctic Peninsula ice cores capture
- 119 changes in the SAM, ENSO and tropical teleconnections in the western Pacific (Thomas et al., 2008;
- 120 2013; 2015; 2017; Medley and Thomas, 2019). The Palmer core specifically displays a strong
- relationship with the Inter-decadal Pacific oscillation (IPO) (Emanuelsson et al., 2022a). Thus, Palmer

- 122 provides an optimal location to capture the deposition of anthropogenic proxies, due to the
- 123 continent's remote location, involves long-range atmospheric transport and a clear pathway of
- 124 northerly air-mass incursions.

125 The availability of uncontaminated frozen sample with sufficient volume for new analysis

126 An advantage of the Palmer ice core, over many deep ice cores, is that it was drilled without drilling

- 127 fluid. The high-density drilling fluid, used routinely to keep the ice core borehole open, is one of the
- main sources of chemical and biological contamination in ice cores (Alekhina et al., 2018). While
- most Antarctic ice cores which span the mid-20th century were drilled in the 1990s (e.g., Thomas et
- al., 2017), the Palmer core was drilled in 2012 and has therefore not experienced any potential
- degradation or damage associated with long-term storage. A key objective of the AWG was to
 undertake new analysis of anthropogenic proxies, requiring substantial ice volumes. Unlike more
- undertake new analysis of anthropogenic proxies, requiring substantial ice volumes. Unlike mo
 established and heavily sampled Antarctic ice cores, the Palmer ice core has sufficient sample
- volume for new analysis whilst meeting the GSSP requirements to maintain an archive.
- 135

136 Materials and methods

137 Geographic setting of core sites

138 The Palmer ice core was drilled in the Palmer Land region of the Antarctic Peninsula [73.86° S, 65.46°

139 W] (Figure 1). The Antarctic Peninsula ice sheet sits within a 2000 m high mountain chain that

140 extends ~1,300-km northward from continental Antarctica toward the southern tip of South

- 141 America. The ice core was drilled at 1897 m above sea level, close to the ice divide, where horizontal
- 142 ice flow is expected to be minimal. The estimated ice thickness at the drill site is ~1000 m (Fretwell
- et al., 2013). With average 2 m temperatures of -28.5°C, and summer averages of -16.2°C, (ERA5
- reanalysis data) this cold site is unlikely to be affected by surface melting. As one of the highest
- points on the ice sheet, the site is distant from any orographic disturbances, such as mountain peaks or areas of exposed rock. The closest rock outcrop is a small nunatak approximately 35 km from the
- drill site. The closest areas of human activity are the summer only stations of Sky Blu (1435 m a.s.l.),
- ²¹⁰ ²¹⁰ ²¹⁰ ¹⁴⁸ ²¹⁰ ¹⁴⁸ ²¹⁰ ¹⁴⁸ ²¹⁰ ¹⁴⁸ ¹⁴⁸ ²¹⁰ ¹⁴⁸ ¹⁴⁸
- to the northwest which has been used intermittently since 1961 CE. The closest year-round research
- 150 station, Rothera (4 m a.s.l.), is over 700 km to the north, which began operation in 1975 CE. Thus,
- 151 the influence of local human activity or light industry is expected to be minimal at this remote
- 152 location, especially prior to the 1960s.

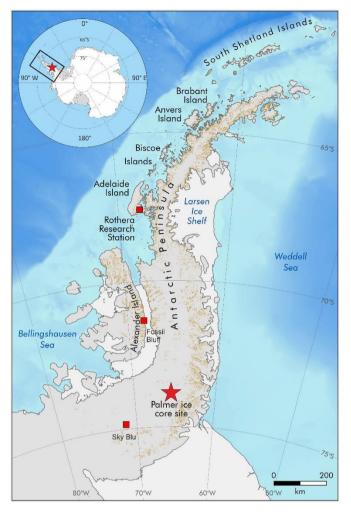


Figure 1. Map of the Antarctic Peninsula showing the Palmer ice core location (red start), and the
 location of research stations. Insert map of the Antarctic content, highlighting the Antarctica

156 Peninsula (black rectangle). Copyright Laura Gerrish, BAS.

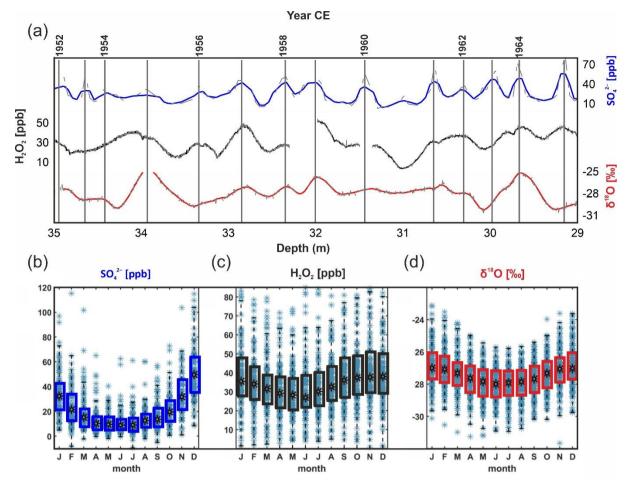
157 Field collection of core and sampling

The ice core was drilled over six days between the $24^{th} - 29^{th}$ December 2012. It was drilled using the 158 British Antarctic Survey (BAS) electromechanical dry drill, which operates in a dry borehole without 159 160 drill fluid. The final drilling depth was 133.45m, drilled in approximate 1 m sections. The cores were cut to 85 cm length, using a cross-saw, and wrapped in pre-labelled layflat tubing. The cores were 161 162 weighed and measured in the field before being loaded into insulated boxes, with polystyrene 163 inserts to prevent core damage during transport. The boxes were stored in a trench in the field, where the atmospheric temperature remained below -20°C. The ice was transported via Twin Otter 164 aircraft to Rothera research station, loaded into a -25°C refrigerated container and shipped to BAS 165 166 for analysis and long-term storage. The ice samples, and remaining archive, are kept at -25°C in a monitored, temperature-controlled freezer facility in the dedicated BAS ice core laboratories in 167 168 Cambridge, UK.

169 Chronological controls

- 170 A suite of chemical and isotopic species analysed using continuous flow analysis (CFA), using
- 171 longitudinal ice sections (32 x 32 mm). Ice samples are loaded onto a heated melt head (in a -25°C
- 172 freezer) and the sample stream of water and gas, from trapped air bubbles, are separated in a

- 173 debubbler before being pumped into a series of analytical instruments (Grieman et al., 2022). The
- bespoke CFA system comprises Fast Ion Chromatography (FIC), Dionex ICS-3000, to measure major
- anions sulphate [SO₄²⁻], nitrate [NO3-], chloride [Cl-], and methane sulphonic acid [MSA-]. The FIC
- data acquisition is one 2.63 cm sample every 4 cm, thus providing an average depth resolution of 4
- 177 cm (Grieman et al., 2022). Hydrogen peroxide $[H_2O_2]$ was measured using a FIAlab photomultiplier
- tube-fluorescence detector (PMT-FL), connected to the CFA. The estimated resolution for the fluorescence detection is ~1.4 +/- 0.5 cm (Grieman et al., 2022). Water isotopes (δ^{18} O) were
- fluorescence detection is ~1.4 +/- 0.5 cm (Grieman et al., 2022). Water isotopes (δ¹⁸O) were
 measured using a Picarro analyzer (L2130-i) operating in a custom-made continuous-flow setup.
- 181 Internal standards, calibrated to the V-SMOW2/Standard Light Antarctic Precipitation 2 (V-
- 182 SMOW2/SLAP2) scale, were used to calibrate the isotope records. The average depth resolution for
- 183 δ^{18} O is ~4 cm.
- 184 The number of data points per year is dependent on the ice depth and annual snow accumulation
- rate. During the 30-year interval surrounding the proposed GSSP (1940-1970), the average snow
- accumulation was 43 ± 27 cm snow per year. Providing an average of 10 (±6) data points per year for
- 187 major ions and δ^{18} O, and 30 (±10) data points per year for H₂O₂.
- 188 The ice core was dated using annual layer counting, based on seasonal deposition of chemical and
- isotopic species (Fig. 2). Clear seasonal cycles are observed in sulphate [SO₄²⁻], which peaks during
- 190 the austral summer (November January) when marine productivity is highest, and hydrogen
- 191 peroxide (H_2O_2), a photochemical species which peaks during the summer solstice. $\delta^{18}O$ also displays
- a seasonal cycle, although with a weaker amplitude than $[SO_4^{2-}]$ and H_2O_2 (Fig. 2d) (Emanuelsson et
- al., 2022b). The annual layers were counted manually, assigning years where a clear peak was
- observed in at least two of the chronological markers SO_4^2 and H_2O_2 , or $\delta^{18}O$. Years with only one
- 195 clear marker, or during unavoidable data gaps (e.g., core breaks), were marked as uncertain.
- 196 The age-scale is independently verified by identifying volcanic $[SO_4^{2-}]$ peaks, classified as $[SO_4^{2-}]$
- 197 values which exceed two standard deviations (2σ) above the mean (Fig. 3a). To account for the
- natural variability in background [SO₄²⁻] we apply a running 200 data-point (approx. 10-years) mean
- 199 and standard deviation (Fig. 3b).



201

Figure 2. Annual layer counting. (a) Seasonal cycles in sulphate $[SO_4^{2^-}]$ (blue), hydrogen peroxide [H₂O₂] (black), and stable water isotopes $[\delta^{18}O]$ (red) from 1952-1965 CE. High resolution data for all species shown in grey dashed curves, solid lines represent a 3-point running mean (7-point running mean for $[H_2O_2]$). Seasonality plots modified from Emanuelsson et al., (2022b) for (b) $[SO_4^{2^-}]$ (blue), (c) $[H_2O_2]$ (black), and (d) $[\delta^{18}O]$ (red). Monthly averaged values for each month, the coloured boxed indicating the 25th and 75th percentiles.

208 Anthropocene proxies

209 Radioisotopes

- 210 Discrete samples were cut to annual resolution, spanning the period 1930 to 1998 CE. The surface
- area of each annual sample was 11.4 cm², and the sample volumes ranged between 64 mL 430 mL.
- 212 This reflects the varying snow accumulation during this period (1930-1998 CE), which can range
- between 0.11 and 0.57 m of water equivalent per year. Subsequently, the annual samples were
- combined to two-yearly resolution due to the extremely low sample concentrations detected. The
- 215 average sample volume (1930-1998 CE) was 459 ± 366 ml.
- 216 All samples were analysed in the GAU-Radioanalytical Laboratories at the University of
- 217 Southampton. Samples were spiked with Pu-²⁴² recovery tracer, transferred into PTFE beakers, and
- evaporated to dryness. Samples underwent a series of acid treatments, with the residues
- 219 evaporated to dryness after each treatment. First residues were dissolved using concentrated
- 220 hydrofluoric acid (HF) solution (2x10 ml), followed by nitric acid (HNO₃) (3x20ml), and finally boiled
- 221 undercover in 40ml concentrated hydrochloric acid (HCl) for 2 hours. After this 2g of boric acid was
- added and the samples were boiled again until complete boric acid dissolution (2hrs).

- 223 Solutions were evaporated to dryness, re-dissolved in 30ml 1M HNO₃ and boiled again under cover
- until a completely clear solution was obtained. 10mg of iron (Fe) carrier was added and ²³⁹⁺²⁴⁰Pu was
- 225 co-precipitated with Fe(OH)₃ using concentrated ammonia solution. Obtained precipitates were
- dissolved in HCl and acid molarity was adjusted to 9M. Samples were loaded onto anion exchange
- columns (1x5cm), previously conditioned with 9M HCl. After the load solution passed the columns,
 columns were washed with further 30ml of 9M HCl followed by 50ml of 8M HNO₃ and again 10 ml of
- 228 columns were washed with229 9M HCl.
- 230 Finally, ²³⁹⁺²⁴⁰Pu was stripped from the columns using 9MHCl/NH₄I (ammonium iodide) solution into
- 231 clean beakers. Solutions were evaporated to dryness with 5ml conc HNO3 added to remove excess
- iodide. Thin alpha-spectrometric sources were prepared using electrodeposition from diluted
- HCl/oxalate solution. Prepared sources were counted for ²³⁸Pu and ²³⁹⁺²⁴⁰Pu using Alpha Octete
- 234 spectrometers with PIPS detectors and spectra analysed using Maestro 32 software. Detection limits
- 235 were ca. 0.5 uBq/g.

236 Spheroidal carbonaceous fly-ash particles (SCPs)

- 237 SCPs are only produced by the high temperature, industrial combustion of coal-series and oil fuels
- 238 (Rose 2015). They are morphologically distinct under the light and scanning electron microscope
- 239 making them unambiguous indicators of deposition from these sources (Rose 2008). Two methods
- 240 were applied to identify the presence of SCPs in the ice. The first approach used discrete samples cut
- from the archive section of the Palmer ice core. The second approach used scanning electron
- 242 microscopy of filters of melt water collected as part of the CFA analysis.
- Method 1. Annual samples were provided between 1900 to 1998 CE. The ice core sample volumes
 provided ranged between 64 mL to 430 mL, and SCP analysis was conducted on every sample (total
 of 98) down-core. The surface area of each annual sample was 11.4 cm².
- Each ice core water sample, representing a single year, was filtered through a glass microfibre (GF/C) 246 247 filter. The filtering apparatus was rinsed with deionised water between samples. The filter papers 248 were then analysed for SCPs following a method adapted from Rose (1994), involving dissolution of 249 the filter paper using hydrofluoric acid followed by a treatment with hydrochloric acid to remove any 250 precipitate formed from the filter (Rose, 1994). Between acid treatments, the samples were washed 251 with distilled water, centrifuged, and decanted. This was repeated twice as a final washing step to 252 remove any remaining acid. SCPs are composed mostly of elemental carbon and are chemically 253 robust and undamaged by the acid process. A known fraction of the final suspension was evaporated 254 onto multiple coverslips and mounted onto microscope slides. The number and sizes of SCPs on the 255 coverslips were counted using a light microscope at x400 magnification using criteria for SCP 256 identification (Rose, 2008). Analytical blanks were included in duplicate for each sample batch, and
- 257 no SCPs were observed in these blanks.
- 258 Method 2. The second approach used the melt water produced while processing ice samples on the 259 BAS CFA system (Grieman et al., 2022). Melt water from the CFA waste lines was collected in new 260 and sealed low-density polyethylene (LDPE; Nalgene™) bottles. Melt water represents the effective 261 deposition of snow and particles over a 6.53 cm² surface. Samples were collected at annual 262 resolution between 2011and 1980 CE (33 samples), and between 5 and 8-year resolution between 263 1900 and 1979 CE (16 samples). Sample volumes ranged between 44 mL and 927 mL. Meltwater 264 from each sample was then filtered through 13 mm diameter, 1.0 µm pore size Whatman™ 265 Polycarbonate membrane filters, inside clean polypropylene Swinnnex[™] filter holders. Each filter 266 was then mounted onto an aluminium stub for analyses on a scanning electron microscope (SEM) in the Earth Sciences Department at the University of Cambridge. Filters were imaged on a Quanta-267

- 268 650F using back scattered electrons (BSE) on a low-pressure mode. Each filter was imaged at x800
- 269 magnification for SCPs identification, following the analysis strategy presented in (Tetzner, 2021).
- 270 SCPs were identified, based on their characteristic morphological and textural features. Analytical
- 271 blanks were included every 15 samples in the filtration process. No SCPs were observed in the
- 272 blanks.

273 *Methane*

274 The Palmer methane (CH₄) record was measured using a continuous flow analysis (CFA) (Stowasser 275 et al., 2012; Grieman et al., 2022). The melted water stream (with an average melt rate of 3-5 276 cm/min) is pumped through three steps of gas extraction; 1) a debubbler, where gravity takes away 277 excess water, 2) a hydrophobic membrane, which separates gas bubbles from the water completely, 278 and 3) a Nafion dryer, which absorbs water vapor on a molecular level. The dry gas sample stream is 279 measured continuously with the commercially available laser spectrometer Picarro G2301 using 280 wavelength-scanned cavity ring-down spectroscopy. The effective CH₄ stratigraphic resolution is 1.5-17.5 cm. The instrumental CFA CH₄ uncertainty is 10 ppb. The raw CH₄ mixing ratio was calibrated to 281 282 NOAA gas standards (https://gml.noaa.gov/ccl/airstandard.html) to obtain absolute values and 283 corrected for solubility in the meltwater stream. NOAA primary air standards (405.8 ppb and 869.1 284 ppb [CH₄] calibrated against the WMO (World Meteorological Organization) X2004A reference scale 285 Dlugokencky et al., 2005) were measured before and after the CFA analysis providing a slope of 286 1.034 and intercept of 3.708. The percentage of dissolved gas was calculated daily based on an 287 experiment mimicking a melted water stream by mixing a working standard gas ([CH₄] = 606.97 ppb) 288 and deionized water. An average factor to increase the CH₄ mixing ratio is 8.75%.

- The age of the CH₄ (or any other gas) is not equal to the age of the ice at a certain depth. The so-
- called gas-ice age difference (or Δ Age) (Lemieux-Dudon et al., 2010) relates to the atmospheric air
- trapping when the glacier ice forms. Pores between grains close at a certain depth (typically
- between 50 to 70 m in ice sheets but can be down to 120 m (Buizert et al., 2021)), based on the
- snow and firn (granular snow that has not yet been compressed into ice) densification process.
- The gas age at the depth that the pores lock-in (56.8 m for Palmer) is 0 years, because the air is
- exchanged in the opened pores with the atmospheric air on the surface. Throughout the lock-in zonebubbles close steadily, and the air defuses until the close-off depth. This occurs at 62.8 m, where
- 297 the Δ Age is 17 years. The Δ Age is unique to each ice core site and often assumed to be constant for
- 298 calculation convenience. One can identify ΔAge either by firn densification modelling (see e.g.
- Buizert et al., 2021) or by synchronizing with existing atmospheric gas records (Blunier et al., 2007).
- 300 In this study, synchronisation to the Law Dome CH₄ record (Rubino et al., 2019) is adopted, matching
- the absolute values and slopes of the two records. The ΔAge was verified using the community firn
- 302 model (Stevens et al., 2020) implemented using a varying snow accumulation rate, ranging from 0.17
- to 0.33 m w.e. yr⁻¹ based on measured density, annual layer depths (Emanuelsson et al. 2022a) and
- an annual average surface air temperature estimate of -28.5°C (ERA 5 reanalysis). The Δ Age from the community firn model approach ranges from 112 to 162 years, in agreement with the Δ Age estimate
- 306 based on gas synchronisation of 90 to 140 years.

307 Numerical analysis

308 Change points in CH₄ detected using a Bayesian Estimator of Abrupt change, Seasonal change, and

- 309 Trend (BEAST) (Zhao et al., 2019). It is a decomposition algorithm with built-in Bayesian model
- 310 averages. It allows identification of a time stamp of a long-term trend change in a noisy time series.

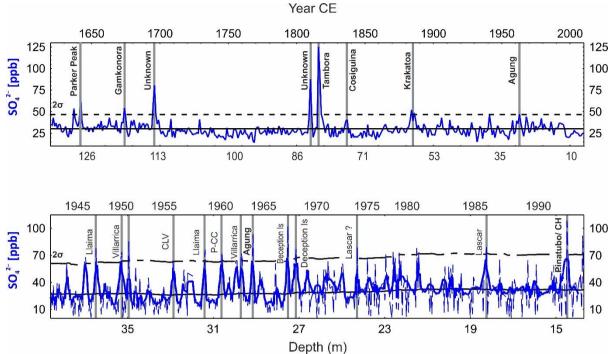
- For CH₄, the change-point analysis was run using 5-yearly averages (to reflect the mixing in the firn
- column) and presented with an estimated error of +/- 5 years.

313 Results

314 Chronology

The Palmer ice core covers 391 years, 1621–2011 CE, and is one of the oldest records from the

- 316 Antarctic Peninsula. The annual layer counted age scale is verified using the dates of known volcanic
- eruptions that have been detected in ice cores from both Antarctic and Arctic ice cores (Sigl et al.,
- 2013). Six distinct peaks are observed in the annual averaged $[SO_4^{2-}]$ (Fig 3a) corresponding to
- volcanic eruptions documented in the WAIS divide ice core (Sigl et al., 2013). This includes Parker
- 320 peak (1640), Gamkonora (1673), Tambora (1815), Cosiguina (1835), Krakatoa (1883), and Agung
- (1963). Two additional peaks are assigned to eruptions of unknown sources (1694 and 1809), which
 have been widely documented and identified in other Antarctic and Arctic sites (Emanuelsson et al.,
- 2022b). The chronological error is established by evaluating the absolute age differences of volcanic
- horizons in Palmer, compared with documented ages in the WAIS divide ice core (Sigl et al., 2013).
- 325 The maximum age difference is 6-months and thus our chronological error is ± 0.5-year.
- 326 The 1963 eruption of Agung, Indonesia, provides the closest tie-point for the proposed GSSP. While
- 327 the annual average $[SO_4^{2-}]$ does not exceed the threshold of 2σ above the mean, the threshold is
- exceeded in the raw (~4 cm) data, at a depth of 29.2 m corresponding to 1964 (Fig 3b). The 1991
- 329 eruption of Pinatubo, Philippines, is another well-documented eruption visible in many polar ice
- 330 cores (Sigl et al., 2013). The threshold of 2σ is also only exceeded in the raw data, as a distinct [SO₄²⁻
- 331] peak at 14.5 m corresponding to 1992 (Fig 3b). A delay of between one and three years is expected
- between the volcanic eruption and the deposition of $[SO_4^2]$ at an Antarctic ice core site (Cole-Dai et
- 333 al., 1997).
- A number of smaller, more proximal, eruptions provide additional age constrains during the mid-20th
- century (3b) (Global Volcanism Program., 2022). Deception island, located in the South Shetland
- islands to the north of the Antarctic Peninsula is known to have been active between 1968 and 1970
- 337 with a volcanic explosivity index (VEI) of 3. This is coincident with multiple $[SO_4^{2-}]$ peaks that exceed
- 2σ (and 3σ) above the running 200-point average between 29.5 and 27.09 m (1968-1969 CE). Two
- peaks in $[SO_4^{2-}]$ at 36.5 m and 34.9 m most likely correspond to documented eruptions of Llaima
- 340 (1945 CE) and Villarrica (1948/1949 CE), two stratovolcanoes from southern Chile with documented
- VEI 3 eruptions (Global Volcanism Program, 2022). These peaks are used to constrain the lower
- 342 portion of the GSSP period.
- 343



344

345 Figure 3. Volcanic reference horizons in the Palmer chronology plotted on a depth (bottom axis) and age-scale (top axis). (a) Annual average [SO₄²⁻], highlighting volcanic peaks (grey vertical lines) 346 identified in other Antarctic and bi-polar ice cores. Solid horizontal line indicates record average and 347 348 dashed horizontal line indicates 2σ above the record average. (b) Raw (~4 cm resolution) [SO₄²⁻] 349 between 1942 and 1993 CE, highlighting both mid-latitude (Agung and Pinatubo, bold) and Southern 350 Hemisphere eruptions (Global Volcanism Program., 2022). Solid vertical lines indicate peaks 351 exceeding 2σ above a 200-point running average (solid black curve), with corresponding dated 352 volcanic eruptions exceeding VEI 3. Cerro Hudson (CH), Carran-Los Venados (CLV) and Puyehue-

353 Cordon Caulle (P-CC).

354

355 Radioisotopes

356 The ²³⁹⁺²⁴⁰Pu activity concentrations are presented in Figure 4 and 5. The first evidence of ²³⁹⁺²⁴⁰Pu in

the Palmer ice core is at 37.16 m depth, corresponding to years 1945-1946 CE. This constitutes the

358 lowest concentration (0.39 μ B/g) in the Palmer record. The concentrations of ²³⁹⁺²⁴⁰Pu at Palmer

increase after 1945, through a subsidiary maximum in the early 1950s, reaching an absolute

360 maximum (1.38 μ B/g) at 30.48 m by 1960-1961.

361 The low concentrations are consistent with previous ²³⁹⁺²⁴⁰Pu measured in Antarctic ice cores

362 (Arienzo et al., 2016), which are considerably lower than concentrations measured in Greenland ice

363 cores or Alpine ice cores (Gabrieli et al., 2013) or other mid-latitude archives (Waters et al., 2015).

Temporal trends are also broadly similar to those reported previously (e.g., Arienzo et al. 2016), with

two exceptions, in the date of first detection of Pu in the ice core, and post-1980 concentration

trends. In previous studies, the first detected Pu in Antarctic ice cores was 1952 CE, while in Alpine

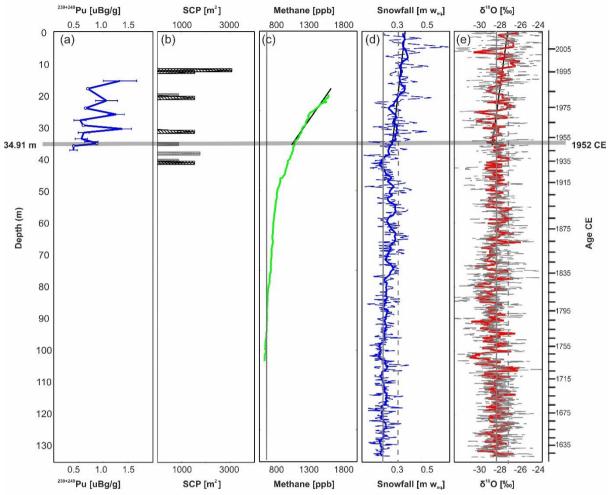
367 cores it was as late as 1954 CE. The earlier detection in the Palmer ice core may reflect the different

analytical approaches, which has previously been measured using Inductively Coupled Plasma –

Sector Field Mass Spectrometer (ICP-SFMS) (e.g., (Gabrieli et al., 2013). However, the proximity to
 the test sites (surrounding the Pacific region) may also explain the earlier detection on the Antarctic

Peninsula. Back-trajectory analysis indicates that the Pacific sector of the Southern Ocean is the

- dominant source region for the 5-day air-parcels reaching Palmer (Thomas and Bracegirdle, 2015).
- 373 An apparent secondary increase in ²³⁹⁺²⁴⁰Pu in 1990 (albeit with low precision, with concentrations
- 374 close to detection limits), if real, may be a result of increased aerosol or dust supply, and likely
- 375 reflects local processes rather than any regional fallout process.



376

377 Figure 4. Anthropogenic proxies in the Palmer ice core in the context of the last 391 years. (a) Plutonium (²³⁹⁺²⁴⁰Pu) [µBg/g] (solid), with error bars, presented as 2-yearly resolution. Open circles 378 379 indicate data below detection limit. (b) SCPs [m²] determined using discrete sampling (black bars, annual resolution) and continuous sampling (grey bars, 1–5-year resolution). Auxiliary climatological 380 381 records (c) (CH_4 [ppb], (d) annual snowfall (meters of water equivalent) (dashed line) with running 382 decadal mean (thick solid curve) and (e) δ^{18} O (‰) at 4cm resolution (grey dashed curve) and annual averages (solid red curve). All data plotted on a depth scale (left axis), with age-scale shown on right 383 384 axis. The proposed GSSP is highlighted in the grey line corresponding to depth 34.91 m (1952 CE).

386 Novel materials

Fly-ash (SCPs). A total of five SCPs were identified using method 1 (discrete sampling), with a further
 six SCPs identified using method 2 (continuous meltwater). These eleven particles constitute the first

evidence that SCPs have been deposited in Antarctica (Rose, 2015), and to our knowledge are the

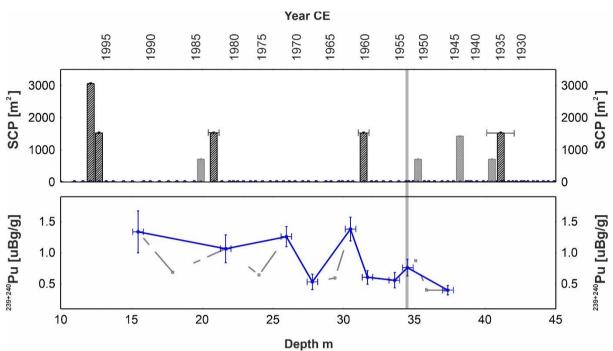
390 first dated record of deposition in an Antarctic ice core. The SCP record is presented as a

391 concentration (m²) in Figure 4 & 5. The first SCP observed using method 1 is in the ice layer

392 corresponding to 1936 CE (40.48 m). For the continuous method (methods 2) the first SCP is

identified in the sample centred on 41.12 m depth, which incapsulates years 1930-1937 CE.

394



395

Figure 5. Anthropogenic proxies in the Palmer ice core between 1930-1998 CE. (a) SCPs [m²]
 determined using discrete sampling (black bars, annual resolution) and continuous sampling (grey
 bars, 1–5-year resolution). Blue dots indicate samples analysed where no SCPs were found. (b)
 Plutonium (²³⁹⁺²⁴⁰Pu) [µBg/g], measured at bi-annual resolution from 1946 to 1991 CE (grey dashed
 curve). Values which exceed the limit of detection shown in blue (presented in Fig. 4). Error bars in
 the x-axis denote depth resolution and error in the y-axis denote analytical uncertainty. Vertical grey
 line denotes the proposed GSSP horizon at 34.91 m (1952 CE).

403 *Geochemical proxies for climate*

- 404 **Stable water isotopes.** δ^{18} O are commonly used to reconstruct past surface temperature from
- Antarctic ice cores (e.g., Stenni et al., 2017). The stable water isotope record from Palmer displays
- 406 evidence of seasonal cycles, ranging from -33.12 ‰ (winter) to -20.51‰ (summer). When converted
- to decadal averages, the values from the most recent decade (2001-2010 CE) are the most
- isotopically enriched (-26.82%), thus suggesting that temperatures during the first decade of the
- 409 21^{st} century were warmer than any decade since 1621 CE. δ^{18} O has been increasing at a rate of 0.22
- 410 % per decade since 1950 CE (Figure 4). This is consistent with other ice cores from the Antarctic
- Peninsula, which display an isotopic warming during the late 20th century (Thomas et al., 2013;
- 412 Stenni et al 2017; Thomas and Tetzner, 2017), consistent with observations.
- 413 Snow accumulation. The amount of snowfall (snow accumulation) per year is determined as the sum
- of precipitation, sublimation, evaporation, and melt (Thomas et al., 2017). Snow accumulation is
- 415 measured using the distance between chronological peaks (SO_4^{2-} and H_2O_2) and converted to metres
- 416 of water equivalent per year (m w_{eq} yr⁻¹) based on the measured density. The record is corrected for
- 417 ice thinning and horizontal flow using the Nye model, which assumes thinning is proportional to
- 418 burial and is appropriate to use in the upper 10-15% of the ice sheet (Nye, 1963).

- 419 The snow accumulation increases from a decadal average of 0.24 m w_{eq} yr⁻¹ at the beginning of the
- 420 record (1621-1630 CE) to a decadal average of 0.38 m $w_{eq}\,yr^{-1}$ in the final decade (2001-2010 CE).
- 421 Snowfall has been increasing at a rate of 1.5 cm per decade since 1950 CE (Figure 4). This increase is
- 422 consistent with other Antarctic Peninsula ice cores (Thomas et al., 2015, Porter et al., 2016) and is
- 423 part of a regional trend, which began in the ~1920s (Thomas et al., 2015; 2017). Snow accumulation
- 424 variability in the Antarctica Peninsula is driven by changes in tropical sea surface temperatures,
- regional sea ice conditions (e.g., (Thomas et al., 2015; Porter et al., 2016; Thomas et al., 2017)) and large scale modes of atmospheric circulation, most notably the SAM (Medley and Thomas, 2019).
- 427 However, the underlying trend in snow accumulation can also be explained by the observed
- 428 atmospheric warming (Medley and Thomas, 2019).

429 Methane.

- 430 The oldest CH₄ measurement corresponds to approximately 1719 CE (+/- 5 years) with a
- 431 concentration of 645 ± 10 ppb. Concentrations of CH_4 increase throughout the record, with a marked
- 432 acceleration during the 20^{th} century (Figure 4). The concentrations of CH₄ have doubled by 1969 CE.
- The concentration at the lock-in depth (56.8 m) is 1839 ± 10 ppb, which corresponds with modern
- CH_4 concentration in the atmosphere. At the pore close-off depth (62.8 m) the concentration is 1511
- 435 ± 10 ppb, which in terms of gas age is approximately 1977 CE. The corresponding concentration in
- the Law Dome ice core in 1977 CE is 1476 ± 5 ppb (Rubino et al., 2019). The earliest observational
- monthly data, from air collected in glass flasks at the Palmer Station in Antarctica (Lan et al., 2022)
 from NOAA Global Monitoring Laboratory, began in January 1983 with a CH₄ concentration of 1557
- 439 ppb.
- 440 Change point analysis identifies two significant changes in the 5-yearly averaged CH₄ below the
- 441 critical depth (1977 CE). The change-points correspond to 1958 and 1883 CE, with probabilities of
- 442 99% and 82% respectively. We emphasize that due to the gradual gas trapping and residence time in
- the atmosphere (9.1 \pm 0.9 years, (Prather et al., 2012)), the CH₄ change points cannot be interpreted
- 444 as an exact year. However, the record suggest that the most significant decadal change detected in
- the 264-year CH_4 record is centred on 1958 (+/- 5 years) CE.

446 Discussion

- 447 The Palmer ice core meets the requirements of a GSSP site. The ice layers are of adequate thickness
- to support global correlation, with an accurate chronology independently verified using well-dated
- volcanic horizons. The Agung eruption of 1963 provides a well-dated reference horizon observed in
- 450 other Antarctic and Greenland ice cores (Sigl et al., 2013) in close proximity to the GSSP depth. The
- 451 Palmer location is also sensitive to volcanic [SO₄²⁻] deposition from South American volcanos, with
- 452 six $[SO_4^2]$ peaks in the 20-year window surrounding the GSSP (1952) coincident with dated Chilean
- 453 eruptions (Fig. 3)(Global Volcanism Program, 2022). Snowfall is continuous, with small variations in
- 454 seasonal deposition (Thomas and Bracegirdle, 2015), ensuring the record is not seasonally biased.
- 455 The risk of reworking is small, with redistribution of surface snow estimated at just 5% (van Lipzig et
- al., 2004). The ice layers, and anthropogenic proxy data they contain, are continuous and unaffected
- 457 by tectonic and sedimentary movements, or metamorphism. For this ice core specifically, the high
- elevation (1897 m) and southerly latitude (74°S) ensure that the site is not at risk of surface melting.
- 459 Even under extreme future warming scenarios, the proposed marker for the onset of the
- 460 Anthropocene is 34.91 m below the surface.
- 461 Antarctica is a unique location, governed by the Antarctic Treaty as a continent for peaceful
- 462 purposes, freedom of scientific investigation and scientific observations and results that shall be

exchanged and made freely available. The Protocol on Environmental Protection designated
Antarctica as a "natural reserve" in 1998, ensuring that future scientific exploration of the Palmer
site is not at risk of exploitation or political changes. The Palmer ice core site is easily accessible by
Twin Otter aircraft from several international research stations on the Antarctic Peninsula, and from
other Antarctic gateways (e.g., South America, South Africa, New Zealand, and Australia). Thus,

despite its remote location, future access for repeat drilling is feasible, achievable, and uniquely

469 protected. The Palmer ice core used in this study is stored at a government funded research facility

- in the United Kingdom, ensuring safe, long-term storage and accessibility for the public and
- 471 academics.

472 The Palmer ice core provides clear evidence that anthropogenic proxies, in the form of SCPs and 473 radionuclides (Pu), have been deposited in Antarctica. Both proxies have no known natural source 474 but are evident in the Palmer ice core from the mid-twentieth century onwards. SCPs have been 475 recorded in the records of coastal mainland and sub-Antarctic lakes sediments (Rose et al., 2012), 476 and in near-shore marine sediments (Martins et al., 2010). However, the SCP data presented here 477 are the first for Antarctic ice and demonstrate for the first time that these markers of heavy industry 478 (high temperature combustion) reached this more remote part of the Antarctic continent as early as 479 1936 CE. The most likely source region of these SCPs is the South American or Australian continents, 480 where coal powered energy industry was delayed relative to the United Kingdom and other nations 481 (Rose and Appleby, 2005). However, the low analytical detection and transport mechanisms may

also in part explain the delayed appearance relative to other global locations.

The presence of ²³⁹⁺²⁴⁰Pu in the samples corresponding to 1945/46 CE indicate that even the earliest bomb tests, which began in July 1945 CE, are detected in the Palmer ice core. The earlier tests were fission weapons, with fallout in the lowest layers of the atmosphere (Aarkrog, 2003). Despite the extremely low concentrations, the detection of ²³⁹⁺²⁴⁰Pu cannot be attributed to any natural source

487 or emission scenario. Tropospheric transport of volcanic ash from South American and mid-latitude

488 volcanoes has been observed reaching West Antarctica just 2-3 weeks following an eruption

489 (Koffman et al., 2017). Thus, we hypothesize that Palmer's location in the Western Antarctic

490 Peninsula, with strong teleconnections with the tropical Pacific (e.g. (Thomas et al., 2013)),

facilitated the tropospheric transport of ²³⁹⁺²⁴⁰Pu to the ice core. Making it possible to detect the fall-

492 out from the first explosions in 1945/46 CE.

493 It was not until after 1952 CE, and the first thermonuclear tests, that stratospheric transport was 494 sufficient to produce a global signal detectable at other polar (Arienzo et al., 2016) and mid-latitude 495 ice core sites (Gabrieli et al., 2011; Gabrieli et al., 2013). While ash layers in ice cores can be used to 496 demonstrate rapid tropospheric transport, the sulphate emitted from explosive volcanic eruptions 497 can often take several years to be detected in Antarctic ice (Koffman et al., 2017). This may explain the delayed deposition in of ²³⁹⁺²⁴⁰Pu at other polar and mid-latitude locations. While the first 498 detection of ²³⁹⁺²⁴⁰Pu occurs in 1945/1946 CE, the first recorded peak in values (significantly above 499 500 the detection limit) corresponds to 1952/53 CE. This is consistent with the first thermonuclear tests

501 and is a globally identifiable feature (Waters et al., 2015).

502 The highest concentrations of ²³⁹⁺²⁴⁰Pu (1960/61 CE) are coincident with the largest nuclear test (Tsar 503 Bomba) in October 1961 CE. The radioactive debris from such large thermonuclear explosions can 504 remain in the stratosphere for between 15-18-months (Zander and Araskog, 1973). Thus, despite the 505 extremely distant source region for this test (Novaya Zemlya, Arctic Russia), the sufficiently long 506 time-window for global stratospheric transport is feasible. This is also supported by other well 507 documented stratospheric transport and deposition of particles and chemical species in the 508 Antarctic ice core record (e.g., volcanic eruptions) (Sigl et al., 2013; Koffman et al., 2017). The brief

- 509 drop to lower concentrations during years 1963-1967 CE, may reflect the reduction in nuclear testing
- 510 following the partial test ban treaty in 1963 CE. A sharp decrease in deposition corresponding to
- 511 1967 CE has previously been observed in an ice core from the Alps (Gabrieli et al., 2011),
- 512 demonstrating the sensitivity of the ice core record to detect changes in ²³⁹⁺²⁴⁰Pu emissions. By 1970
- 513 CE, the concentrations of ²³⁹⁺²⁴⁰Pu returned to the 1960/61 CE levels, despite most of the testing
- being moved underground. This may reflect the delayed and ongoing stratospheric ²³⁹⁺²⁴⁰Pu
- 515 deposition. The analogous stratospheric transport of sulphate aerosols has been documented in the
- 516 Antarctic ice core record several years after the eruption took place (Koffman et al., 2017). ²³⁹⁺²⁴⁰Pu
- 517 is still detected as recently as 1990/1991 CE.
- 518 Both SCPs and ²³⁹⁺²⁴⁰Pu are distinct markers of the Anthropocene, with no known natural source. The 519 long half-life of ²³⁹Pu (24,110 years), together with the high weathering resistance of the SCPs, mean
- 520 they will both be preserved in the ice as a *near*-perpetual marker. Thus, the presence of both could
- 521 be used as a primary stratigraphic marker that defines the lower boundary of the Anthropocene.
- 522 However, based on the globally synchronous detection of ²³⁹⁺²⁴⁰Pu corresponding to the first
- 523 thermonuclear tests, we propose that ²³⁹⁺²⁴⁰Pu be the primary marker at this site. The SCPs provide a
- 524 clear secondary marker for the Palmer GSSP, visible under magnification.
- 525 In addition to the clear evidence of Anthropogenic proxies (SCPs and ²³⁹⁺²⁴⁰Pu), the auxiliary
- 526 climatological records confirm that the Palmer site has experienced significant climate and
- 527 environmental change since the 1950s. The observed isotopic warming, and positive trend in snow
- 528 accumulation, since 1950 is consistent with regional observations (e.g., Thomas et al., 2017; Stenni
- et al., 2017), suggesting warmer atmospheric surface temperatures and increased precipitation. CH₄
- 530 provides a unique record of past atmospheric conditions that place this proposed GSSP site in a
- global context. Although produced by a variety of natural processes, the acceleration of CH_4 in the
- atmosphere in recent centuries has been linked to changes in land use, landfills, agricultural activity,
 industrialisation, and coal mining (Kirschke et al., 2013), the latter directly linked to the SCP record,
- as a marker for coal combustion. As a more potent greenhouse gas than carbon dioxide, CH_4
- accounts for more than one-quarter of the radiative imbalance (IPCC, 2013) and plays a major role in
- 536 driving present and future climate. CH₄ does not represent a spike, or a single stratigraphic point,
- and is thus not proposed as a GSSP marker. However, change point analysis identifies a statistically
- 538 significant marker centred around 1958 CE (+/- 5 years), which is broadly coincident with the first
- recorded ²³⁹⁺²⁴⁰Pu peak (1952/1953 CE). Thus, we have included CH₄ in this study to provide a unique
 global context to support this GSSP proposal.
- 541 **Table 1.** Summary of anthropogenic proxies detected in the Palmer ice core and their ranking as a
- 542 GSSP marker. Presented with the year they are first detected (²³⁹⁺²⁴⁰Pu and SCPs), their peak
- 543 concentration (²³⁹⁺²⁴⁰Pu and SCPs) and the proposed year to mark the onset of the Anthropocene.

Proxy/ record	First detected	Peak (CE)	GSSP (CE)	Depth	GSSP ranking
	(CE)			(m)	
²³⁹⁺²⁴⁰ Pu	1946	1960	1952	34.91	Primary
SCPs	1936	1996	1936	40.73	Secondary

- 545 To conclude, we propose the Palmer ice core as a candidate for the Global Boundary Stratotype
- 546 Section and Point (GSSP) to mark the onset of the Anthropocene. Based on the first peak in ²³⁹⁺²⁴⁰Pu,

our primary marker, we propose that the onset of the Anthropocene was 1952 CE observed at a

548 depth of 34.91 m.

- 549 While ice cores have been used as previous GSSP sites (Walker et al., 2009), to date, there are no
- 550 GSSP sites in Antarctica. Thus, if this site is selected as the GSSP for the Anthropocene we propose it
- be named the "Antarctican". The anthropogenically driven mass loss from the Antarctic ice sheet,
- observed since the mid-20th century, is predicted to produce 5 cm of global mean sea level rise by
- 553 2100 (DeConto et al., 2021). Thus, it seems especially fitting that this new epoch be named for the
- continent that will arguably play the largest role in governing the planet's future habitability.
- 555

556 Data availability

- 557 The Palmer data is available from the UK Polar Data Centre (UKPDC)
- 558 https://doi.org/10.5285/b3eca350-79aa-49b2-bd6b-ffee86ad6559, and
- 559 <u>https://doi.org/10.1017/jog.2021.75.</u>
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