



**HAL**  
open science

# Variability of marine $^{14}\text{C}$ reservoir ages in the Southern Ocean highlighting circulation changes between 1910 and 1950

Martine Paterne, Elisabeth Michel, Virginie Héros

► **To cite this version:**

Martine Paterne, Elisabeth Michel, Virginie Héros. Variability of marine  $^{14}\text{C}$  reservoir ages in the Southern Ocean highlighting circulation changes between 1910 and 1950. *Earth and Planetary Science Letters*, 2019, 511, pp.99-104. 10.1016/j.epsl.2019.01.029 . hal-02462774

**HAL Id: hal-02462774**

**<https://hal.science/hal-02462774>**

Submitted on 24 Jun 2021

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 **VARIABILITY OF MARINE <sup>14</sup>C RESERVOIR AGES IN THE SOUTHERN OCEAN**  
2 **HIGHLIGHTING CIRCULATION CHANGES BETWEEN 1910 AND 1950**

3 Martine Paterne<sup>1</sup>, Elisabeth Michel<sup>1</sup>, Virginie Héros<sup>2</sup>

4 <sup>1</sup>Laboratoire des Sciences du Climat et de l'Environnement, LSCE/IPSL, CEA-  
5 CNRS-UVSQ, Université Paris-Saclay, Domaine du CNRS, F-91198 Gif-sur-Yvette

6 <sup>2</sup>Muséum d'Histoire Naturelle de Paris, Institut de Systématique & Evolution,  
7 ISyEB (UMR 7205 CNRS/UPMC/MNHN/EPHE) 43 rue Cuvier 75231 Paris France

8

9

10 **Abstract**

11 Recent changes in the Southern Ocean overturning circulation have been attributed  
12 either to natural variability or to anthropogenic forcing. The <sup>14</sup>C analyses of mollusk shells  
13 from Kerguelen Islands evidence a high decadal variability of the sea surface radiocarbon  
14 reservoir ages in the southern South Indian ranging from 832 to 1005 (±30) years between  
15 1910 and 1930. Such high variability is common to the different sectors of the Southern  
16 Ocean, (southern South Pacific and Atlantic), at inter-annual and decadal timescales between  
17 1901 and 1950. Notably, a decrease of  $\Delta^{14}\text{C}$  of ~40‰ (increased R of ~290 years) occurred  
18 between 1901 and 1940, followed by an increase of ~30‰ between 1940 and 1958. Using a  
19 simple box model of ocean-atmosphere <sup>14</sup>CO<sub>2</sub> exchange, this aging is attributed to increased  
20 upwelling of cold and <sup>14</sup>C-depleted subsurface seawaters due to intensified poleward Southern  
21 Westerlies. The sea surface <sup>14</sup>C aging is indeed well correlated to a positive phase of the  
22 Southern Hemisphere Annular Mode and to a cooling of the sea surface temperatures in the

23 Southern Ocean between 1901 and 1940. These pre-1950 sea surface  $^{14}\text{C}$  changes highlight  
24 the importance of natural changes in wind intensities and ocean dynamics.

25

26 **Keywords:** Radiocarbon, Reservoir ages, Kerguelen Is., Southern Westlies- driven  
27 upwelling, Southern Ocean,

28

29

## 30 **1. Introduction**

31 The Southern Ocean (SO) is an important component of the global oceanic circulation  
32 with the redistribution of water masses between the Pacific, Atlantic and Indian Oceans by the  
33 Antarctic Circumpolar Current (ACC) (Fig. 1) (Tamsitt et al., 2017 and references therein).  
34 The strong westerlies in the Southern Hemisphere, combined with the Coriolis force, drive the  
35 ACC and a northward Ekman transport, permitting deep waters to upwell up to the surface at  
36 the Antarctic Divergence. In the last few decades, surface westerlies intensified poleward  
37 (Thompson & Solomon, 2002) that may give rise to significant changes in ocean circulation,  
38 carbon and heat uptake and biological productivity (Toggweiler & Samuels, 1995; Le Quéré  
39 et al., 2007; Sallée et al., 2010, 2012; Waugh et al., 2013; de Vries et al., 2017).

40 Significant changes of ocean ventilation in the Southern Ocean, such as the increase in  
41 the age of the circumpolar deep waters (CDW), were inferred from the analysis of  
42 chlorofluorocarbon-12 (CFC12) from early 1990s to 2000s and transport model simulations  
43 (Waugh et al., 2013). Similarly, using a global data assimilation ocean inverse model, and a  
44 carbon cycling model, De Vries et al. (2017) found an enhanced upper-ocean overturning  
45 circulation in 1990s relatively to 1980s and increased outgassing of natural CO<sub>2</sub>. Both  
46 findings from analysis of CFC-12 and from inverse modeling are consistent with the expected  
47 increased northward Ekman drift and intensified upwelling resulting from westerlies  
48 intensification (Hall & Visbeck, 2002; Waugh et al., 2013; De Vries et al., 2017). Wind  
49 intensity is associated to a zonally symmetric structure in the atmospheric pressure gradient  
50 between the southern mid-latitudes and the polar regions of the SO, referred to as the  
51 Southern Annular Mode (SAM) (Thompson & Wallace, 2000; Thompson et al., 2000). In a  
52 positive phase the Westerlies, associated with higher than normal sea level pressures at the  
53 mid-latitudes and lower than normal at the high latitudes, are reinforced poleward, driving  
54 increased northward Ekman drift and intensified upwelling (Hall & Visbeck, 2002).

55 South of the Polar Front (PF), the sea surface reservoir ages (R), the difference  
56 between the marine and atmospheric  $^{14}\text{C}$  ages at the same calendar age, are highly variable  
57 ranging from ~1000 to ~1500 years. They are based on the  $^{14}\text{C}$  dating of solitary corals,  
58 mollusk shells, bones and flesh remains of penguins and seals of known ages, the  $^{14}\text{C}$  of  
59 which representing that of the dissolved inorganic carbon (DIC) in seawater in which they  
60 grow. The values of R reflect the balance between the  $^{14}\text{C}$  production and the ocean-  
61 atmosphere exchange of  $^{14}\text{CO}_2$  as well as ocean dynamics (Stuiver et al., 1986) and their  
62 temporal variability reflect changes among these factors. Particularly, changes in zonal wind  
63 intensities in the Antarctic regions may significantly modify the ocean-atmosphere exchange  
64 and mixing between the sea surface and the  $^{14}\text{C}$ -depleted underlying subsurface waters, and  
65 consequently the sea surface  $^{14}\text{C}$  ages.

66 We present here new values of R estimated from mollusk shells collected between  
67 1910 and 1930 off the Kerguelen *Is.* in the southern South Indian (SSI) where no values of R  
68 exist presently (Fig. 1). They are compared to previous estimates in the southwestern South  
69 Atlantic (SSA) (off Signy *Is.*) and in the southeastern South Pacific (SSP) (off the western  
70 Antarctic Peninsula and in the Ross Sea) in order to better understand the causes of the high  
71 variability of R in the Southern Ocean. We restricted our comparison to biogenic carbonates,  
72 though few, as penguins and seals remains may capture the  $^{14}\text{C}$  content of different water  
73 masses due to the variety of diving depths, often very far from the Antarctic coasts (Biuw et  
74 al., 2007) (supplementary material, Table S1). After a brief description of the main  
75 hydrographic features at all the sample locations, the sea surface  $^{14}\text{C}$  values are compared to  
76 those of DIC from early  $^{14}\text{C}$  measurements of water samples before the nuclear bomb tests  
77 (Broecker et al., 1960; Rafter, 1972) and from the GLODAP reanalyses of the GEOSECS  
78 campaigns in the 70's (Rubin & Key, 2002; Key et al., 2004). They are further compared to  
79 the variations of the SAM index, based on the time series of the sea level pressure difference

80 between the Southern subtropical regions and Antarctica since 1860 (Marshall, 2003; Visbeck,  
81 2009; Fogt et al., 2009; Jones et al., 2009) and to those of the annual sea surface temperature  
82 (SST) anomalies zonally averaged over the latitude bands 50°S-70°S, referred to as the  
83 Southern Ocean centennial variability (SOCV) index (Latif et al., 2013).

84

## 85 **2 Material and methods**

86 Mollusk shells, collected on Kerguelen Is., were obtained from the collection of the  
87 Muséum National d'Histoire Naturelle de Paris (Fig. 1; supplementary material and Table S1).  
88 They are related to epifaunal species of gastropods, including two active predators  
89 *Neobuccinum eatoni* Smith (specimens from collections Rallier du Baty 1909 and Loranchet  
90 1914) and one herbivore, *Nacella kerguelensis* Smith (collection Aubert de la Rue 1931). The  
91 date of collection corresponds to the year of sampling during scientific explorations on  
92 Kerguelen Is. by Rallier du Baty expedition in 1908/1909 and in 1913/1914, and by Aubert de  
93 la Rue in 1931. One epifaunal gastropod (1914) is still presenting tissues. The shells of the  
94 two other specimens are whole and intact, so they did not wear away before being collected,  
95 confirming that they were sampled alive.

96 Methods for radiocarbon analyses were described elsewhere (Tisnerat-Laborde et al.,  
97 2001). Chemical pre-treatment and carbonate hydrolyses were performed at the Gif-LSCE  
98 Radiocarbon Laboratory and graphite targets were prepared and measured at the French  
99 National AMS-ARTEMIS-LMC14 facility (Moreau et al., 2013). Results are expressed in  
100 conventional years BP (Table S1) and the  $\delta^{14}\text{C}$  ‰ is the deviation in parts per thousand (‰)  
101 of a sample's  $^{14}\text{C}/^{12}\text{C}$  ratio corrected for year of growth from that of the absolute standard  
102 activity in 1950 (Stuiver and Polach, 1977).  $R$  is the difference between the measured  $^{14}\text{C}$  age  
103 and the atmospheric  $^{14}\text{C}$  age from the calibration record SHCal 13 in the southern Hemisphere

104 (Hogg et al., 2013), and  $\Delta R$  the difference between the measured  $^{14}\text{C}$  age and the modeled  
105 ocean one using the marine calibration dataset Marine13 (Reimer et al., 2013) at the date of  
106 the collection. Difference between previous published values and those presented in Table S1  
107 are due to the different utilized calibration record.

108

### 109 **3 Oceanographic setting**

110 The Kerguelen plateau (water depth <1000m) is a major topographic barrier where the  
111 eastward flowing ACC subdivides in two parts with the main flows associated to the Sub-  
112 Antarctic Front (SAF) and to the south by the Southern ACC front (SACCF) (Park et al.,  
113 2014) (Fig. 1). Between SAF and SACCF is located the polar front (PF), the path of which is  
114 observed either north or south of the Kerguelen *Is.* (Sokolov and Rintoul, 2009; Kim and Orsi,  
115 2014; Park et al., 2014) and subject to large northward/southward shifts up to 100 km in a few  
116 tens of years (Sokolov and Rintoul, 2009; Kim and Orsi, 2014).

117 Deep waters from the southern Pacific, Atlantic and Indian Oceans enter the ACC  
118 within the Circumpolar Deep Water (CDW) (Orsi et al., 1993, 1995; Whitworth et al., 1994;  
119 Talley, 2013). From the South Atlantic, most of NADW, marked by high salinity in SO,  
120 enters the ACC, where it is modified by mixing with the locally formed deep waters of the  
121 lower core of CDW (LCDW). A small branch of NADW flows eastward directly into the  
122 South Indian upwelling as a component of the less dense deep Indian Water. The Indian and  
123 Pacific Deep Waters move southward to the ACC above NADW and form the upper core of  
124 CDW (UCDW), characterized by an  $\text{O}_2$ -minimum (Orsi et al., 1995; Talley, 2013). UCDW  
125 upwelling is balanced by a northward flow of low salinity that descends from the polar to the  
126 sub-Antarctic front (SAF) to form the Antarctic Intermediate Water characterized by a salinity

127 minimum. LCDW flows poleward and sinks along the continental slope mainly in the  
128 Weddell and Ross Seas and along Adélie Land as the Antarctic Bottom Water.

129         The Signy *Is.* are located in the vicinity of the Drake Passage on the Scotia Ridge  
130 where surface waters result from the mixing of the water bodies from the ACC and from the  
131 northwestern Weddell Sea (Whitworth et al., 1994) (Fig. 1). To the north of the shelf is  
132 located the southern front of the ACC where UCDW and LCDW upwell (Whitworth et al.,  
133 1994; Orsi et al., 1995). To the south, a branch of NADW penetrates CDW in the South  
134 Atlantic east of the Weddell Sea gyre near longitude 0°. Then it flows westward directly  
135 underneath the Antarctic surface water (AASW) and when reaching the Antarctic shelves it is  
136 transformed to the Weddell Sea Bottom Water Shelf waters (Whitworth et al., 1994; Orsi et  
137 al., 1995). From the South Atlantic, another branch of NADW flows eastward toward the  
138 Kerguelen Plateau (Orsi et al., 1993). Over the plateau and upstream Kerguelen *Is.*, AASW  
139 overlay the cold Winter Water (WW) at a water-depth of around 180-200 m (Park et al., 2008;  
140 Bowie et al., 2015). UCDW was observed below at 500m water-depth overlying LCDW  
141 (Bowie et al., 2015). Downstream, UCDW is observed at 600-800m water-depth with LCDW  
142 deeper than 1300m (Park et al., 2008; Quéroué et al., 2015). Over the southern Kerguelen  
143 plateau, UCDW is observed at depth of 400-1400m over LCDW (Park et al., 2008). On the  
144 western part of the Antarctic Peninsula, WW are observed at a water-depth of ~100m below  
145 the cold AASW (Orsi et al., 1995; Moffat et al., 2009). The UCDW intrudes the shelf as  
146 frequent and short events, but not as large-scale intrusions, while the 50-150m thick LCDW is  
147 inflowing to the shelf at depths of approximately 500m and below (Moffat et al., 2009).

148         All the considered samples of biogenic carbonates are located in the Antarctic surface  
149 waters within or south of the ACC (Fig. 1).

150



## 151 4 Results and Discussion

### 152 1) Values of R and $\delta^{14}\text{C}$ off Kerguelen *Is.* and comparison with previous 153 determinations of mollusk shells and seawater $\delta^{14}\text{C}$ in the Southern Ocean

154 Values of R and  $\Delta^{14}\text{C}$  of the biogenic carbonates ( $\Delta^{14}\text{C}_{\text{carb}}$ ) off Kerguelen *Is.* range  
155 from  $832 \pm 31$  years to  $1005 \pm 32$  years and from  $-108.2 \pm 3.2$  to  $-131.9 \pm 3.2$  ‰, respectively,  
156 between 1909 and 1931 (Fig. 2; Table S1). Compared to these, greater R and more depleted  
157  $\Delta^{14}\text{C}_{\text{carb}}$  were measured from mollusk shells off the Antarctic Peninsula and from the eastern  
158 Ross Sea varying from  $1088 \pm 58$  years to  $1326 \pm 40$  years and from  $-136.9 \pm 6$  ‰ to  $-166.8 \pm$   
159  $3.9$  ‰ respectively in the two regions. Values at Signy *Is.* are also greater than the measured  
160 ones off Kerguelen *Is.* at an average of  $1109 \pm 89$  years and  $-146 \pm 9.5$  ‰. The statistical Chi-  
161 square tests indicate that, in each sector, the values of R and  $\Delta^{14}\text{C}_{\text{carb}}$  are incompatible at the  
162 95% confidence level attesting a highly significant variability of  $^{14}\text{C}$  in the Antarctic surface  
163 water.

164 We compared the  $\Delta^{14}\text{C}_{\text{carb}}$  to the natural  $\Delta^{14}\text{C}_{\text{DIC}}$  estimated in their respective regions  
165 (Broecker et al., 1960; Rafter, 1972; Rubin & Key, 2002; Key et al., 2004) (Fig. 3;  
166 supplementary material, Table S2). Mean values of  $\Delta^{14}\text{C}_{\text{DIC}}$  in UCDW and LCDW are  
167 calculated using the temperature, salinity and  $\text{O}_2$  properties defined by Orsi et al. (1995). The  
168 sea surface and UCDW/LCDW values of  $\Delta^{14}\text{C}_{\text{DIC}}$  are similar within the uncertainties in the  
169 southern South Pacific (SSP) along the Antarctic Peninsula and in the Ross Sea and they are  
170 not significantly different from those of Signy *Is.* in the southwestern South Atlantic (SSA).  
171 Values of  $\Delta^{14}\text{C}_{\text{DIC}}$  in SSP and SSA are consequently grouped into a unique sector. The mean  
172  $\Delta^{14}\text{C}_{\text{DIC}}$  are lower in SSP-SSA than in SSI by  $\sim 20$  ‰ for both the sea surface and  
173 UCDW/LCDW. In all the sectors, the values from mollusk shells mimic the high variability

174 of AASW  $\delta^{14}\text{C}_{\text{DIC}}$  and some are close or similar to those of UCDW and LCDW. This is also  
175 featured by the  $\delta^{14}\text{C}_{\text{carb}}$  of the corals from the Ross Sea ice shelves (Hall et al., 2010).

176 At Kerguelen *Is.*, the values of  $\delta^{14}\text{C}_{\text{carb}}$  linearly decrease with a highly significant  
177 regression coefficient ( $r^2=0.98$ ) between 1909 and 1931 (Fig. 2). In the SSP-SSA sector,  
178  $\delta^{14}\text{C}_{\text{carb}}$  significantly decrease linearly between 1917 and 1940 ( $r^2=0.76$ ) while they do not  
179 vary between 1917 and 1953 ( $r^2=0.05$ ), that is partly due to the high inter-annual variability of  
180  $\delta^{14}\text{C}_{\text{carb}}$  of  $\sim 18\%$  at Signy *Is.* at around 1950. This is also related to a well-marked reverse  
181 increasing trend in 1940 up to 1958 ( $r^2=0.73$ ) considering the measured  $\delta^{14}\text{C}_{\text{DIC}}$  in 1958 in the  
182 southern South Atlantic, south of the ACC, and in the Ross Sea where the bomb-produced  $^{14}\text{C}$   
183 effect had a minimum impact (Broecker et al., 1960; Rafter, 1972) (Table S2). Between 1917  
184 and 1940, the equation of the linear regression in SSP is very similar to that of SSI. Moreover,  
185 it is also similar and highly significant ( $r^2=0.87$ ) when adding one value in 1901 from one  
186 coral from the Ross Sea (Hall et al., 2010) and when accounting for the calendar age  
187 uncertainty, which is greater than that of mollusk shells (supplementary material, Fig. S1).  
188 Finally, the negative trends of  $\delta^{14}\text{C}_{\text{carb}}$  in SSI and SSP between 1901 and 1940 have an offset  
189 of  $\sim 20 \pm 7\%$  that is very consistent with the estimated difference of  $\delta^{14}\text{C}_{\text{DIC}}$  between these  
190 sectors (Table S2). Subtracting 20% to the values of  $\delta^{14}\text{C}_{\text{carb}}$  at Kerguelen *Is.* results in a  
191 significant decrease of sea surface  $^{14}\text{C}$  ( $r^2=0.90$ ) between 1901 and 1940. We thus conclude  
192 that the observed decreases of  $\delta^{14}\text{C}_{\text{carb}}$  in the southern South Indian and South Pacific are  
193 robust statistical trends between 1901 and 1940 as well as the increase of  $\delta^{14}\text{C}_{\text{carb}}$  and  
194 between 1940 and 1958. The uncertainty of  $\delta^{14}\text{C}$ , estimated from the square root of the  
195 variance, is  $\pm 7\text{-}9\%$  between 1901 and 1958 that is of the same order than the mean inter-  
196 annual changes of measured  $\delta^{14}\text{C}_{\text{carb}}$  at Signy *Is.*. In addition to the inter-annual variability,  
197 the record of  $\delta^{14}\text{C}$  from the biogenic carbonates and seawater DIC very likely reflects long-  
198 term changes of sea surface  $^{14}\text{C}$  in the Southern Ocean, which are very likely due to common

199 processes acting on the global Southern Ocean sea surface  $^{14}\text{C}$ . Using the values of the slopes  
200 and their uncertainties, the decrease of  $\Delta^{14}\text{C}_{\text{carb}}$  would be of some  $\sim 10 \pm 2\%$  per decade  
201 between 1901 and 1940 and the sea surface  $\Delta^{14}\text{C}$  increase per decade would be of some  $18 \pm$   
202  $5\%$  between 1940 and 1958.

203

## 204 **2) Factors contributing to the variations of sea surface $^{14}\text{C}$ in the Southern Ocean**

206 The inter-annual and decadal changes of sea surface  $\Delta^{14}\text{C}$  in the Southern Ocean are  
207 much greater than the changes of  $^{14}\text{C}$  in the atmosphere. Notably, the sea surface  $^{14}\text{C}$  aging  
208 between 1900 and 1940 is 2 times greater than the contemporaneous aging of the atmosphere  
209 due to the industrial and domestic input of  $^{14}\text{C}$ -depleted fossil fuels, known as the *Suess effect*  
210 (Hogg et al., 2013). Changes of the sea surface  $^{14}\text{C}$  in the Southern Ocean can neither be  
211 attributed to changes in the extent of the sea-ice cover that inhibits the atmosphere-ocean  
212  $^{14}\text{CO}_2$  exchange, nor to changes in the outflow of old freshwaters from subglacial lake brines,  
213 such as in the Ross Sea (Mikucki et al., 2009), as such features do not exist at Kerguelen Is..

214 Changes of the sea surface  $^{14}\text{C}$  in the Southern Ocean may be the result of the  
215 variations of the flux of NADW into the CDW core waters as  $\Delta^{14}\text{C}_{\text{DIC}}$  is higher in NADW at  
216 around  $-100\%$  (Broecker et al., 1960) than in the southern South Indian and Pacific deep  
217 waters (Fig. 3). They may be also caused by changes of wind-induced mixing of sea surface  
218 with the cold and  $^{14}\text{C}$ -depleted subsurface waters from UCDW or LCDW mediated by  
219 changes of either the mixed layer depth (MLD) or Ekman-induced upwelling. Nonetheless,  
220 the greatest wind-induced changes in MLD were observed within and north of the ACC rather  
221 than in the southernmost regions of the Southern Ocean (Sallée et al., 2010; Carranza and  
222 Gille, 2014).

223 We thus estimated the changes of  $R$  due to changes either in the NADW flux into the  
224 Southern Ocean or in upwelling intensities from simplified equations for  $\Delta$  a surface water  
225 box, exchanging with a deep box CDW and the atmosphere (supporting material, Fig. S2).  
226 The flux of NADW into CDW was changed by a factor of 2, as observed during the past 60  
227 years for NADW intensity in the North Atlantic (Bryden et al., 2014; Atkinson et al., 2012).  
228 Augmenting the contribution of NADW from 40% (Ganachaud, 2003; Talley, 2013) to 57%  
229 decreases  $R$  in SO by 25 years, that is much lower than the measured decadal changes of  $\sim$ 80  
230 years. Testing a 40% increase of the upwelling in SO, the sea surface  $^{14}\text{C}$  ages were  
231 augmented by  $\sim$ 100 years. In our simple model, using a sinusoidal change in SO upwelling,  
232 the time lag of the maximum  $^{14}\text{C}$  age response to changes in wind-driven upwelling fluxes is  
233 6 years, but a coupled ocean-atmosphere model would better estimate the impact of winds on  
234 both the amplitude and timing of the oceanic changes of  $^{14}\text{C}$  in SO. The 40% increase in  
235 upwelling at the Antarctic Divergence used in the test is of the same order of magnitude than  
236 that estimated by DeVries et al. (2017), using data assimilation of global oceanographic  
237 tracers (CFC, temperature and salinity) in the 1990s relative to 1980s in an inverse model of  
238 the upper ocean circulation. The impact of increased upwelling is clearly a robust candidate to  
239 account for the long-term sea surface changes of 80 years per decade in the SO.

240 We compared the temporal changes of  $\Delta^{14}\text{C}$  against the 11 year - running mean  
241 filtered records of the SAM index (Visbeck, 2009) and of the SOCV index (Latif et al., 2013)  
242 (Fig. 4). After subtracting 20 ‰ from SSI  $\Delta^{14}\text{C}_{\text{carb}}$  in order to normalize to SSP, the values of  
243  $\Delta^{14}\text{C}$  in the Southern Ocean were interpolated using a 3<sup>rd</sup> order polynomial fit between 1901  
244 and 1958. The changes of  $\Delta^{14}\text{C}$  in the Southern Ocean are significantly correlated with the  
245 variations of SAM ( $r^2=0.67$ ) with a time lag of  $\Delta^{14}\text{C}$  of 6 years and with those of SOCV  
246 ( $r^2=0.94$ ) and a time lag of  $\Delta^{14}\text{C}$  of 12 years. Between 1901 and 1940,  $\Delta^{14}\text{C}$  decreased when  
247 the wind strength increased and SST became colder and between 1940 and 1958 it increased

248 when the wind strength decreased and SST became warmer. The long-term changes of  $\delta^{14}\text{C}$   
249 in the Southern Ocean are thus likely related to the changes of wind driven upwelling  
250 intensities between 1900 and 1958. The poleward increase of westerlies between 1901 and  
251 1940 led to increased upwelling similarly as determined from CFC analyses and inverse  
252 modeling for the 1980s to the 2000s, when poleward Westerlies intensified (Vaugh et al.,  
253 2013; deVries et al., 2017).

254 In the latitudinal bands 50°-70°S, the SST exhibit one cycle of centennial variability  
255 (Latif et al., 2013) (Fig. 4B). Prior to 1901, few values of  $\delta^{14}\text{C}_{\text{carb}}$  exist from paired  $^{14}\text{C}$  and  
256 U/Th dating of corals from Ross Sea Ice Shelves (Hall et al., 2010). The two values of  
257  $\delta^{14}\text{C}_{\text{carb}}$  in 1847 and 1862 are compatible at  $-144.1 \pm 8.4\%$  and significantly more depleted  
258 than that of the coral in 1901, indicating an increase of  $\delta^{14}\text{C}_{\text{carb}}$  prior to 1901 (supplementary  
259 material). Between 1850 and 1958, the  $\delta^{14}\text{C}$  of corals, mollusks shells and seawater DIC from  
260 the Ross Sea, Antarctic Peninsula and southern South Atlantic would vary along a potential  
261 fluctuation of ~80—100 year, that should be examined more thoroughly.

262 The recent changes in wind intensities were attributed either to natural decadal  
263 variability or to anthropogenic forcing due to increased greenhouse gases and ozone depletion  
264 (Thompson and Solomon, 2002; Thompson et al., 2011) or both (Vaugh et al., 2013; DeVries  
265 et al., 2017). Between 1910 and 1940, wind intensities increase markedly such as between  
266 1980 and 2000 (Visbeck, 2009), but the  $\text{CO}_2$  input into the atmosphere increases moderately  
267 by 10ppm (Rubino et al., 2013) while it augments by 45 ppm between 1980 and 2000  
268 (Keeling 1976; Thoning et al., 1989). The changes of  $\delta^{14}\text{C}_{\text{carb}}$  in the Antarctic surface waters  
269 that we linked to fluctuations in wind driven upwelling intensities provide further evidence of  
270 the natural variability of the coupled ocean-atmosphere system at low frequency.

271

## 272 **5 Conclusion**

273 The  $^{14}\text{C}$  analyses of mollusk shells collected on Kerguelen *Is.* evidence a high decadal  
274 variability of the sea surface radiocarbon reservoir ages in the southern South Indian ranging  
275 from 830 to 1005 ( $\pm 30$ ) years between 1909 and 1931. Such high variability of sea surface  
276  $^{14}\text{C}$  is common to the different sectors of the Southern Ocean at inter-annual and decadal  
277 timescales. In addition to inter-annual changes of  $\Delta^{14}\text{C}_{\text{carb}}$  of 18‰ as observed in the  
278 southwestern South Atlantic, a noticeable sea surface decrease of  $\sim 40\%$  (increased R of  $\sim 290$   
279 years) was evidenced between 1900 and 1940, followed by an increase of  $\sim 30\%$  between  
280 1940 and 1958. These pre-1950 variations are large compared to atmospheric variations and  
281 highlight the natural variability of the Southern Ocean reservoir ages.

282 Between 1900 and 1958, the temporal changes of sea surface  $^{14}\text{C}$  in the Southern  
283 Ocean are well correlated to the indices of the wind strength and SST changes of SAM and  
284 SOCV, respectively. Aging of sea surface occurred when SAM changed to a positive phase  
285 and SST became colder between 1901 and 1940, and inversely later up to 1958. We suggest  
286 that wind driven upwelling of cold and  $^{14}\text{C}$ -depleted waters up to the sea surface is the main  
287 factor controlling the observed changes in the sea surface  $^{14}\text{C}$  content in the Southern Ocean.

288 Measurements of the past changes of sea surface  $^{14}\text{C}$  from biogenic carbonates, such  
289 as those between 1840 and 1950, provide one way to approach the low frequency natural  
290 variability of the coupled ocean-atmosphere system. Further detailed  $^{14}\text{C}$  analyses and model  
291 simulations would help in separating the natural and anthropogenic causes of the variability of  
292 the Southern Hemisphere West erlies.

293

## 294 **Acknowledgements**

295 We are greatly indebted to MNHN for the loan of historical malacological material from  
296 Kerguelen Archipelago. We thanked J.R. Toggweiler and an anonymous reviewer for their  
297 useful comments. This work received the financial support of the CEA-CNRS-UVSQ and the  
298 French/Swedish program from the Swedish Research Council (VR-349-2012-6278). Thanks  
299 are due to E. Kaltnecker, C. Noury for technical support and the ARTEMIS-LMC14 team for  
300 graphite targets preparation and AMS-<sup>14</sup>C measurements.

301

302 **References**

- 303 Atkinson, C. P., Bryden, H. L., Cunningham, S. A., King, B. A., 2012. Atlantic transport  
304 variability at 25 N in six hydrographic sections. *Ocean Sci.*, 8, 497–523, doi:10.5194/os-8-  
305 497-2012.
- 306 Biuw, M., Boehme, L., Guinet, C., Hindell, M., Costa, D., Charrassin, J.-B., Roquet, F.,  
307 Bailléul, F., Meredith, M., Thorpe, S., Tremblay, Y., McDonald, B., Park, Y. H., Rintoul,  
308 S. R., Bindoff, N., Goebel, M., Crocker, D., Lovell, P., Nicholson, J., Monks, F., Fedak, M.  
309 A., 2007. Variations in behavior and condition of a Southern Ocean top predator in relation  
310 to in situ oceanographic conditions. *Proc. Nat. Am. Sci.*, 104, 13705-13710.
- 311 Bowie, A. R., van der Merwe, P., Quéroúé, F., Trull, T., Fourquez, M., Planchon, F., Sarthou,  
312 G., Chever, F., Townsend, A. T., Obernosterer, I., Sallée, J.-B., Blain, S., 2015. Iron  
313 budgets for three distinct biogeochemical sites around the Kerguelen Archipelago  
314 (Southern Ocean) during the natural fertilisation study, KEOPS-2. *Biogeosci.*, 12, 4421–  
315 4445.
- 316 Broecker, W.S., Gerard, R., Ewing, M., Heezen, B.C. (1960. Natural Radiocarbon in the  
317 Atlantic Ocean. *J. Geophys. Res.*, 65, 2903-2931.
- 318 Bryden H. L., King B. A., McCarthy G. D., McDonagh, E. L., 2014. Impact of a 30 %  
319 reduction in Atlantic meridional overturning during 2009–2010. *Ocean Sci.*, 10, 683–691,  
320 doi:10.5194/os-10-683-2014.
- 321 Carranza, M. M., Gille, S. T., 2014, Southern Ocean wind-driven entrainment enhances  
322 satellite chlorophyll-a through the summer. *J. Geophys. Res.*, 120, 304–323,  
323 doi:10.1002/2014JC010203.



324 De Vries, T., Holzer, M., Primeau, F., 2017. Recent increase in oceanic carbon uptake driven  
325 by weaker upper-ocean overturning, *Nature*, 542, 214-219, doi:10.10138, nature21068.

326 Fogt, Ryan L., Perlwitz, Judith, Monaghan, Andrew J. Bromwich, David H., Jones, Julie M.,  
327 Marshall G.J., 2009. Historical SAM Variability. Part II: Twentieth-Century Variability  
328 and Trends from Reconstructions, Observations, and the IPCC AR4 Models. *J. Clim.*, 22,  
329 5346-5365. Doi:10.1175/2009JCLI2786.1.

330 Ganachaud, A., 2003. Large-scale mass transports, water mass formation, and diffusivities  
331 estimated from World Ocean Circulation Experiment (WOCE) hydrographic data. *J.*  
332 *Geophys. Res.*, 108(C7), 3213, doi:10.1029/2002JC001565.

333 Hall, A., Visbeck, M., 2002. Synchronous Variability in the Southern Hemisphere  
334 Atmosphere, Sea Ice, and Ocean Resulting from the Annular Mode. *J. Clim.*, 15, 3043-  
335 3057.

336 Hall B. L., Henderson, G.M., Baroni C., Kellogg, T.B., 2010. Constant Holocene Southern-  
337 Ocean <sup>14</sup>C reservoir ages and ice-shelf flow rates. *Earth Plan. Sci. Lett.*, 296, 115-123.

338 Hogg, A.G., Hua, Q., Blackwell, P.G., Niu, M., Buck, C.E., Guilderson, T.P., Heaton, T.J.,  
339 Palmer, J.G., Reimer, P.J., Reimer, R.W., Turney, C.S.M., Zimmerman, S.R.H., 2013.  
340 SHCal 13 Southern Hemisphere calibration, 0–50,000 years cal BP. *Radiocarbon*, 55(4),  
341 1889–1903.

342 Jones, R. L., Fogt, M. Widmann, G. J. Marshall, P. D. Jones, Visbeck, M., 2009. Historical  
343 SAM variability. Part I: Century-length seasonal reconstructions. *J. Clim.*, 22, 5319–5345.

344 Keeling, C.D., Bacastow, R.B., Bainbridge, A.E., Ekdahl, C.A., Guenther, P.R., Waterman,  
345 L.S., 1976. Atmospheric carbon dioxide variations at Mauna Loa Observatory, Hawaii.

346 *Tellus*, 28, 538-551.

347 Key, R. M., Kozyr, A., Sabine, C. L., Lee, K., Wanninkhof, R., Bullister, J. L., Feely, R. A.,  
348 Millero, F. J., Mordy, C., Peng, T.H., 2004. A global ocean carbon climatology: Results  
349 from Global Data Analysis Project (GLODAP). *Global Biogeochem. Cycles*, 18, GB4031,  
350 doi:10.1029/2004GB002247.

351 Kim, Y.S., Orsi, A.H., 2014. On the Variability of Antarctic Circumpolar Current Fronts  
352 Inferred from 1992–2011 Altimetry. *J. Phys. Ocean.*, 44, 3054-3071, DOI: 10.1175/JPO-  
353 D-13-0217.1.

354 Latif, M., Martin, T. Park, W., 2013. Southern Ocean Sector Centennial Climate Variability  
355 and Recent Decadal Trends. *J. Clim.*, 26, 7767-7782.

356 Le Quéré, C., Rödenbeck, C., Buñehuis, E. T., Conway, T.J., Langenfelds, R., Gomez, A.,  
357 Labuschagne, C., Ramonet, M., Nakazawa, T., Metzl, N., Gillett, N., Heimann, M., 2007.  
358 Saturation of the Southern Ocean CO<sub>2</sub> Sink Due to Recent Climate Change. *Science*, 316,  
359 1735-1738, doi :10.1126, science.1136188.

360 Marshall, G.J., 2003. Trends in the Southern Annular Mode from observations and reanalyses.  
361 *J. of Clim.*, 16, 4134–4143.

362 Mikucki, J.A., Pearson, A., Johnston, D.T., Turchyn, A.V., Farquhar, J., Schrag, D.P., Anbar,  
363 A.D., Priscu, J.C., Lee, P.A., 2009. A contemporary microbially maintained subglacial  
364 Ferrous “Ocean”. *Science*, 324, 397-400.

365 Moffat, C., Owens, B., Beardsley, R. C., 2009. On the characteristics of Circumpolar Deep  
366 Water intrusions to the west Antarctic Peninsula Continental Shelf. *J. Geophys. Res.*, 114,  
367 C05017, doi:10.1029/2008JC004955.

368 Moreau, C., Caffy, I., Comby, C., Delqué-Kolif, E., Dumoulin, J., Hain, S., Vincent, J., 2013.  
369 Research and Development of the Artemis <sup>14</sup>C AMS Facility: Status Report. *Radiocarbon*,  
370 55(2), 331-337. doi:10.1017/S0033822200057441.

371 Orsi, A. H., Nowlin W. D. Jr, Whitworth, T., 1993. On the circulation and stratification of the  
372 Weddell Gyre. *Deep-Sea Res. I*, 40, 169-203.

373 Orsi, A. H., Whitworth, T., Nowlin, W. D., 1995. On the meridional extent and fronts of the  
374 Antarctic Circumpolar Current. *Deep-Sea Res. I*, 42, 641–673, doi:10.1016/0967-  
375 0637(95)00021.

376 Park, Y.H., Roquet, F., Durand, I., Fuda, J. L., 2008. Large-scale circulation over and around  
377 the Northern Kerguelen Plateau. *Deep Sea Res., Part II*, 55, 566–581.

378 Park, Y-H., Durand, I., Kestenare, E., Rougier G., Zhou, M., d'Ovidio, F., Cotte, C., Lee, J.H.,  
379 2014. Polar Front around the Kerguelen Islands: An up-to-date determination and  
380 associated circulation of surface/subsurface waters. *J. Geophys. Res.*, 119, 6575–6592, doi:  
381 10.1002/2014JC010061.

382 Quéroué, F., Sarthou, G., Planquette, H.F., Bucciarelli, E., Chever, F., van der Merwe, P.,  
383 Lannuzel, D., Townsend, A.T., Cheize, M., Blain, S., d'Ovidio, F., Bowie, A. R., 2015.  
384 High variability of dissolved iron concentrations in the vicinity of Kerguelen Island,  
385 Southern Ocean). *Biogeosci. Discuss.*, 12, 231–270.

386 Rafter, T., 1972. Carbon-14 variations in Nature, Part 3 measurements in the south Pacific  
387 and antarctic oceans. *New Zealand J. Sci.*, 11, 551-589.

388 Reimer, P.J., Bard, E., Bayliss, A., Beck, J.W., Blackwell, P.G., Bronk Ramsey, C., Buck,  
389 C.E., Cheng, H., Edwards, R.L., Friedrich, M., Grootes, P.M., Guilderson, T.P., Haffidason,  
390 H., Hajdas, I., Hatté, C., Heaton, T.J., Hoffmann, D.L., Hogg, A.G., Hughen, K.A., Kaiser,

391 K.F., Kromer, B., Manning, S.W., Niu, M., Reimer, R.W., Richards, D.A., Scott, E.M.,  
392 Southon, J.R., Staff, R.A., Turney, C.S.M., van der Plicht, J., 2013. Int Cal 13 and Marine 13  
393 radiocarbon age calibration curves 0–50,000 years cal BP. *Radiocarbon*, 55(4), 1869–1887.

394 Rubin, S.I., Key, R.M., 2002. Separating natural and bomb produced radiocarbon in the  
395 ocean: the potential alkalinity method. *Global Biogeochem. Cycles*, 16, 4, 1105,  
396 doi:10.1029/2001GB001432

397 Rubino, M., Etheridge, D. M., Trudinger, C. M., Allison, C. E., Battle, M. O., Langenfelds, R.  
398 L., Steele, L. P., Curran, M., Bender, M., White, J. W. C., Jenk, T.M., Blunier, T., Francey,  
399 R.J., 2013). A revised 1000 year atmospheric  $\delta^{13}\text{C}$   $\text{CO}_2$  record from Law Dome and  
400 South Pole, Antarctica. *J. Geophys. Res. Atmosph.*, 118, 8482–8499,  
401 doi:10.1002/jgrd.50668.

402 Sallée, J.B., Speer, K. G., Rintoul, S. R., 2010. Zonally asymmetric response of the Southern  
403 Ocean mixed-layer depth to the Southern Annular Mode. *Nat. Geosci.*, 3, 273-  
404 279 DOI :10.1038/NGEO812.

405 Sallée, J.B., Matear, R.J., Rintoul, S.R., Lenton, A., 2012. Localized subduction of  
406 anthropogenic carbon dioxide in the Southern Hemisphere oceans. *Nat. Geosci.*,  
407 doi :10.1038/NGEO1523.

408 Schlitzer, R., Ocean Data View, <https://odv.awide>, 2018.

409 Sokolov, S., Rintoul, S. R., 2009. Circumpolar structure and distribution of the Antarctic  
410 Circumpolar Current fronts: 2. Variability and relationship to sea surface height. *J.*  
411 *Geophys. Res.*, 114, C11019, doi:10.1029/2008JC005248.

412 Stuiver, M., Polach, H.A., 1977. Discussion. Reporting of  $^{14}\text{C}$  data. *Radiocarbon*, 19, 355-363.

413 Stuiver, M., Pearson, G.W., Braziunas, T.F., 1986. Radiocarbon age calibration of marine  
414 samples back to 9000 cal yr BP. *Radiocarbon*, 28, 980-1021.

415 Talley, L.D., 2013. Closure of the global overturning circulation through the Indian, Pacific  
416 and Southern Oceans: schematics and transports. *Oceanography*, 26, 80-97.

417 Tamsitt, V., Drake, H.F., Morrison, A.K., Talley, L.D., Dufour, C.O., Gray, A.R., Griffies,  
418 S.M., Mazloff, M.R., Sarmiento, J.L., Wang, J., Weijer, W., 2017. Spiraling pathways of  
419 global deep waters to the surface of the Southern Ocean. *Nat. Com.*, 8, 172, DOI:  
420 10.1038/s41467-017-00197-0.

421 Thompson, D.W.J., Wallace, J.M., 2000. Annular mode in the extratropical circulation, Part I:  
422 Month-to-month variability. *J. Clim.*, 13, 1000–1016.

423 Thompson, D.W.J., Solomon, S., 2002. Interpretation of recent Southern Hemisphere climate  
424 change. *Science*, 296, 895–899.

425 Thompson, D.W.J., Wallace, J.M., Hegerl, G.C., 2000. Annular mode in the extratropical  
426 circulation. Part II: Trends. *J. Clim.*, 13, 1018-1036.

427 Thompson, D.W.J., Solomon, S., Kushner, P.J., England, M.H., Grise, K.M., Karoly, D.J.,  
428 2011. Signatures of the Antarctic ozone hole in Southern Hemisphere surface climate  
429 change. *Nat. Geosci.*, doi :10.1038/NGEO1296.

430 Thoning, K.W., Tans, P.P., Komhyr, W.D., 1989. Atmospheric carbon dioxide at Mauna Loa  
431 Observatory 2. Analysis of the NOAA GMCC data, 1974-1985. *J. Geophys. Res.*, 94,  
432 8549-8565.

433 Tisnéat-Laborde, N., Poupeau, J.J., Tannau, J.F., Paterné, M., 2001. Development of a semi-

434 automated system for routine preparation of carbonate sample. *Radiocarbon*, 43 (2A), 299-  
435 304.

436 Toggweiler, J. R., Samuels, B., 1995. Effect of Drake Passage on the global thermohaline  
437 circulation. *Deep-Sea Res. I*, 42, 477–500.

438 Visbeck, M., 2009. A station-based southern annular mode index from 1884 to 2005. *J. Clim.*,  
439 22, 940–950.

440 Waugh, D. W., Primeau, F., DeVries, T., Holzer, M., 2013. Recent Changes in the Ventilation  
441 of the Southern Oceans. *Science*, 339, 568-570.

442 Whitworth, T. III, Nowlin Jr, W. D., Orsi, A.H., Locarnini, R.A., Smith, S.G., 1994. Weddell  
443 Sea Shelf Water in the Bransfield Strait and Weddell-Scotia Confluence. *Deep-Sea Res. I*,  
444 41, 629-641.

445

446 **Figures Captions**

447 Fig. 1: Map of the Southern Ocean from Ocean Data View (R. Schlitzer, 2018) with the  
448 location of mollusks shells (black stars), corals (white stars) and water samples stations  
449 from the GEOSECS campaigns (numbered circles) and from early cruises (lettered  
450 circles) in Broecker et al. (1960) (B) and Rafter (1972) (R). Hydrographic fronts are  
451 from Orsi et al. (1995): SAF: sub-Antarctic front; PF: polar front; SACC: Southern  
452 front of the ACC; Bdy: southernmost boundary of the ACC. The lower and higher  
453 values of R in the Southern South Indian (SSI), South Atlantic (SSA) and South Pacific  
454 (SSP) are also indicated (supplementary information in text section 1 and Table S1).

455

456 Figure 2: Radiocarbon ages (A), reservoir ages (B) and  $\delta^{14}\text{C}_{\text{carb}}$  (‰) (C) as a function of year  
457 A.D. of mollusk shells from Kerguelen *Is.* (squares), Antarctic Peninsula (black circles)  
458 and Ross Sea (grey circle), Signy *Is.* (triangles) (Table S2) and from Ross Sea corals  
459 (open circles) (Hall et al., 2010). Measured seawater  $\delta^{14}\text{C}_{\text{DIC}}$  in 1958 from the Ross Sea  
460 (dotted circles) (Rafter, 1972) and from the southern South Atlantic in the Weddell Sea  
461 (white triangle) (Broecker et al., 1960) are also shown on C. Between 1901 and 1940,  
462 linear regression equations and coefficients of correlation are shown for values of  
463 Kerguelen *Is.* (SSI); for shell values of the Antarctic Peninsula and Ross Sea (SSP1)  
464 and for shells and coral values of these two last regions (SSP2). Between 1940 and 1958,  
465 the linear regression equations and coefficients of correlation derive from values of  
466  $\delta^{14}\text{C}_{\text{carb}}$  and seawater  $\delta^{14}\text{C}_{\text{DIC}}$  excluding the most  $^{14}\text{C}$ -depleted value at -170‰  
467 (supplementary material). Uncertainties on  $\delta^{14}\text{C}_{\text{carb}}$  ( $1\sigma$ ) for SSI (grey lines), SSP1  
468 (black lines) and SSP2 (dotted lines) are represented prior to 1940, as well as between  
469 1940 and 1958 for SSP(3) (black lines) (see text).

470 Fig. 3: (A) Comparison of  $\delta^{14}\text{C}_{\text{carb}}$  and  $\delta^{14}\text{C}_{\text{DIC}}$  in SSI, SSA and SSP. Values of  $\delta^{14}\text{C}_{\text{carb}}$  and  
471 uncertainties ( $1\sigma$ ) are reported for mollusk shells from Kerguelen *Is.* (SSI), Signy *Is.*  
472 (SSA), Antarctic Peninsula (AP) and Ross Sea (RS) (Table S2) and from corals in the  
473 Ross Sea (Hall et al., 2010) (SSP) with the same symbols than in Figure 2. Values of  
474  $\delta^{14}\text{C}_{\text{DIC}}$  at the sea surface (plus), in UCDW (cross) and LCDW (double cross) are from  
475 early  $^{14}\text{C}$  measurements of water samples before the nuclear bomb tests (B\*: Broecker  
476 et al., 1960; R\*: Rafter, 1972) and from GLODAP reanalyses of natural  $^{14}\text{C}$  from  
477 stations (St) of the GEOSECS campaigns in the 70's (Rubin & Key, 2002; Key et al.,  
478 2004). The mean values of  $\delta^{14}\text{C}_{\text{DIC}}$  and standard deviations are underlined for sea  
479 surface (grey box) and UCDW-LCDW (dark grey box). Analytical errors and standard  
480 deviations of the means are reported at  $1\sigma$ . Analytical errors of UCDW and LCDW, not  
481 drawn for graphic readability, range between 7 to 10 % as an average (Broecker et al.,  
482 1960; Rafter, 1972; Rubin & Key, 2002; Key et al., 2004). Sea surface  $\delta^{14}\text{C}_{\text{DIC}}$  noted  
483 DP (Drake Passage) is from the northern edge of the ACC (supplementary material  
484 Table S2).

485

486 Fig. 4: Comparison of the changes of  $\delta^{14}\text{C}$  (same symbols than in Fig. 2) with the annual  
487 variations of SAM as a function of time (Year) (A). The dotted (annual) and dark grey  
488 (11yr running mean) records are from Visbeck (2009). Thin black line refers to records  
489 from Marshall (2003) and light grey line to Fogt et al. (2009) and Jones et al. (2009).  
490 Records are normalized for graphic convenience and correspond to 11yr running means.  
491 (B) Same comparison with the annual (thin black) and 11yr running mean (bold grey)  
492 variations of SOCV (Latif et al., 2013). The thick black line in A and B represents a  
493 third order polynomial fit between the values of  $\delta^{14}\text{C}$ , once subtracting 20‰ from the



494  $\delta^{14}\text{C}$  from Kerguelen *Is.* (dotted squares) in order to normalize to SSP (see text). A  
495 sinusoidal curve of a 100 yr period (dotted line) is also shown in (B).

496

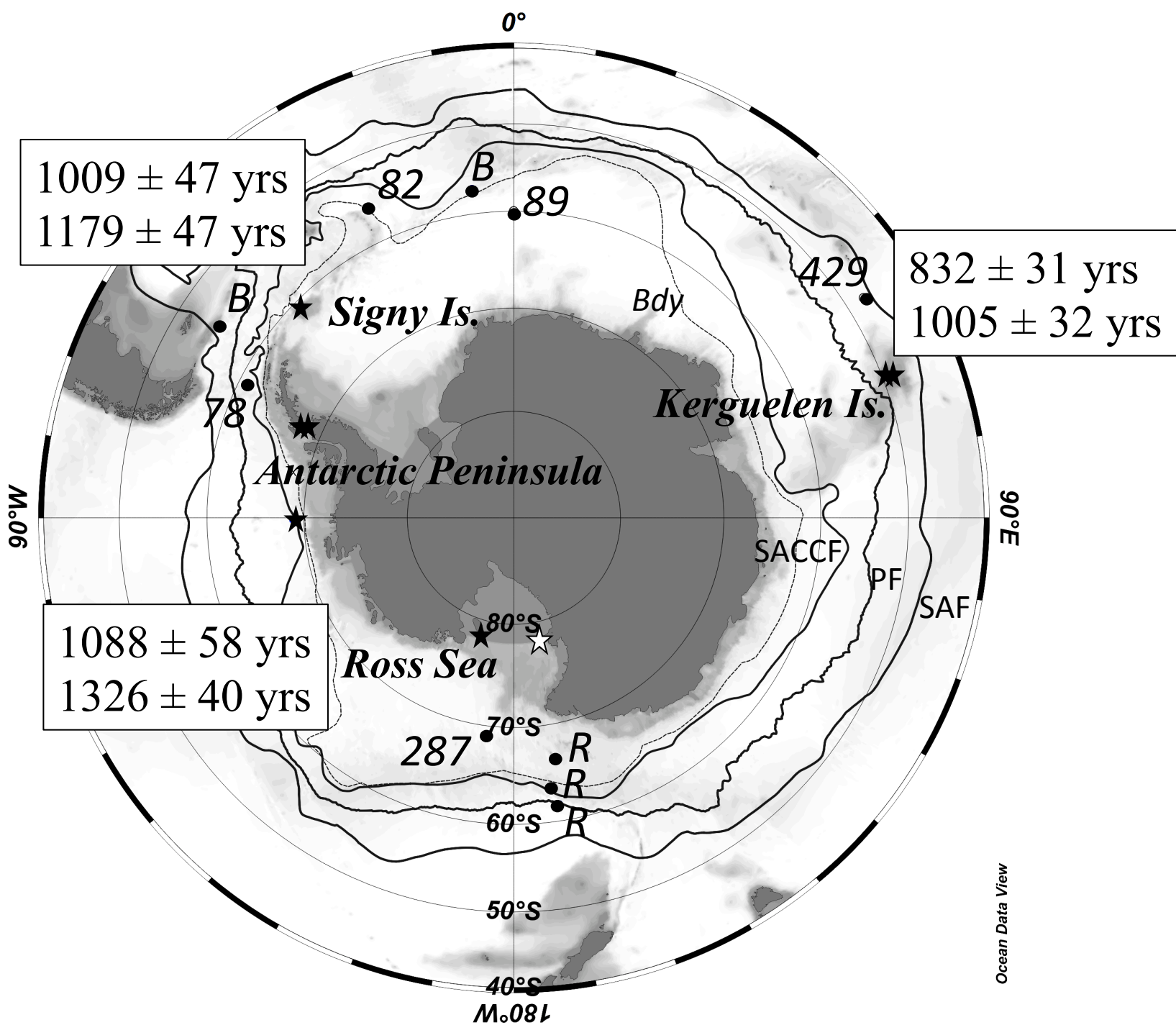


Figure 1

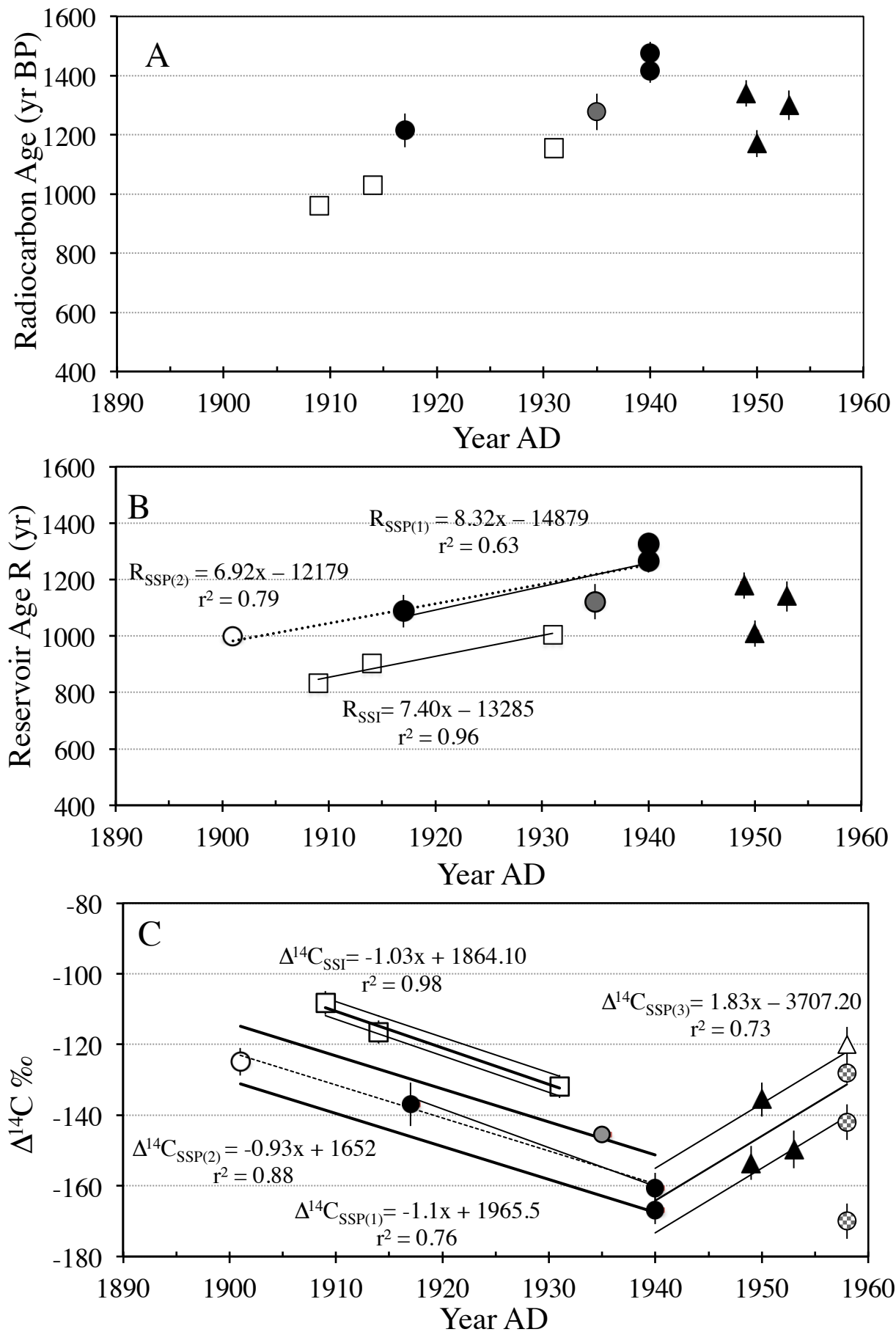


Fig 2

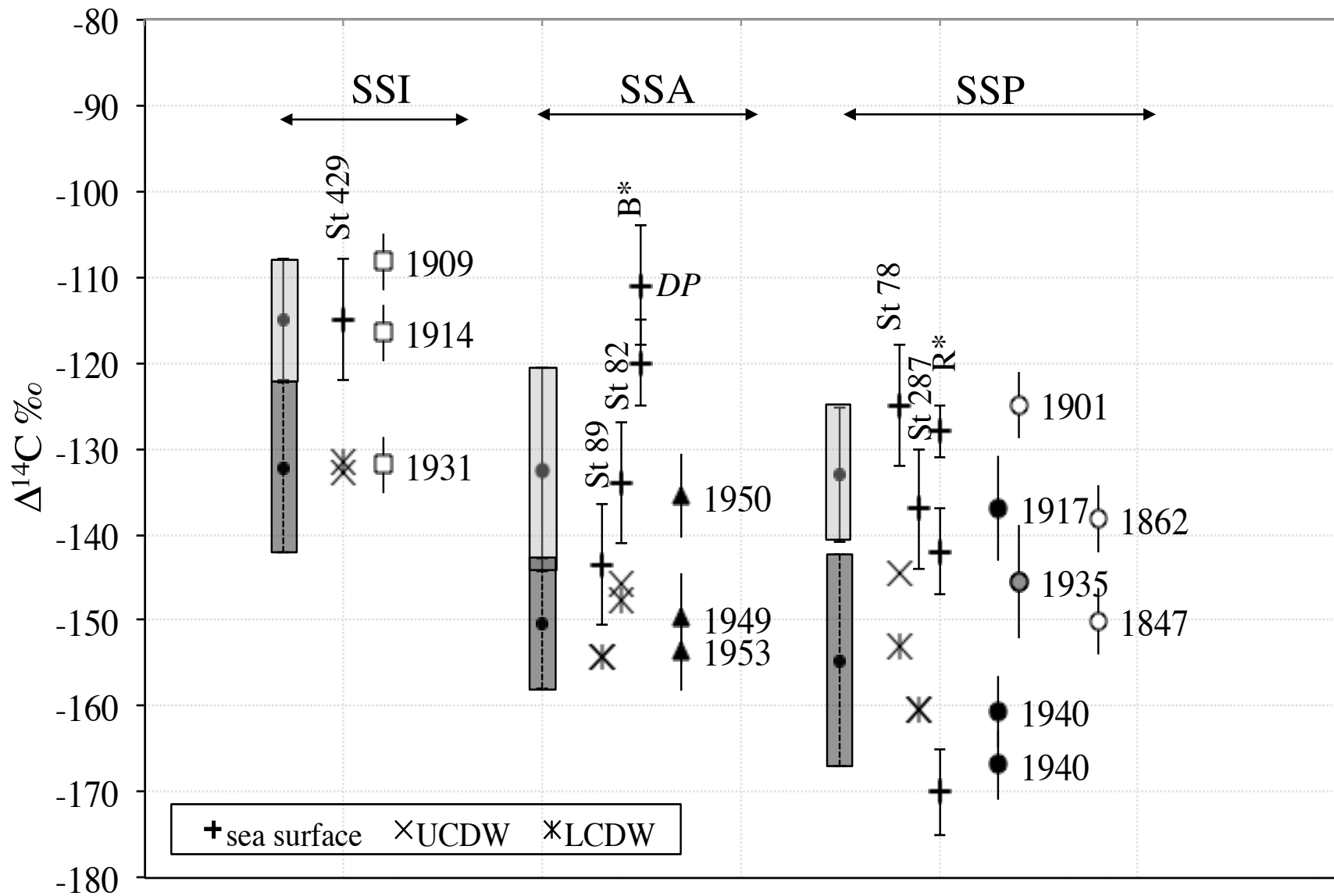


Fig. 3

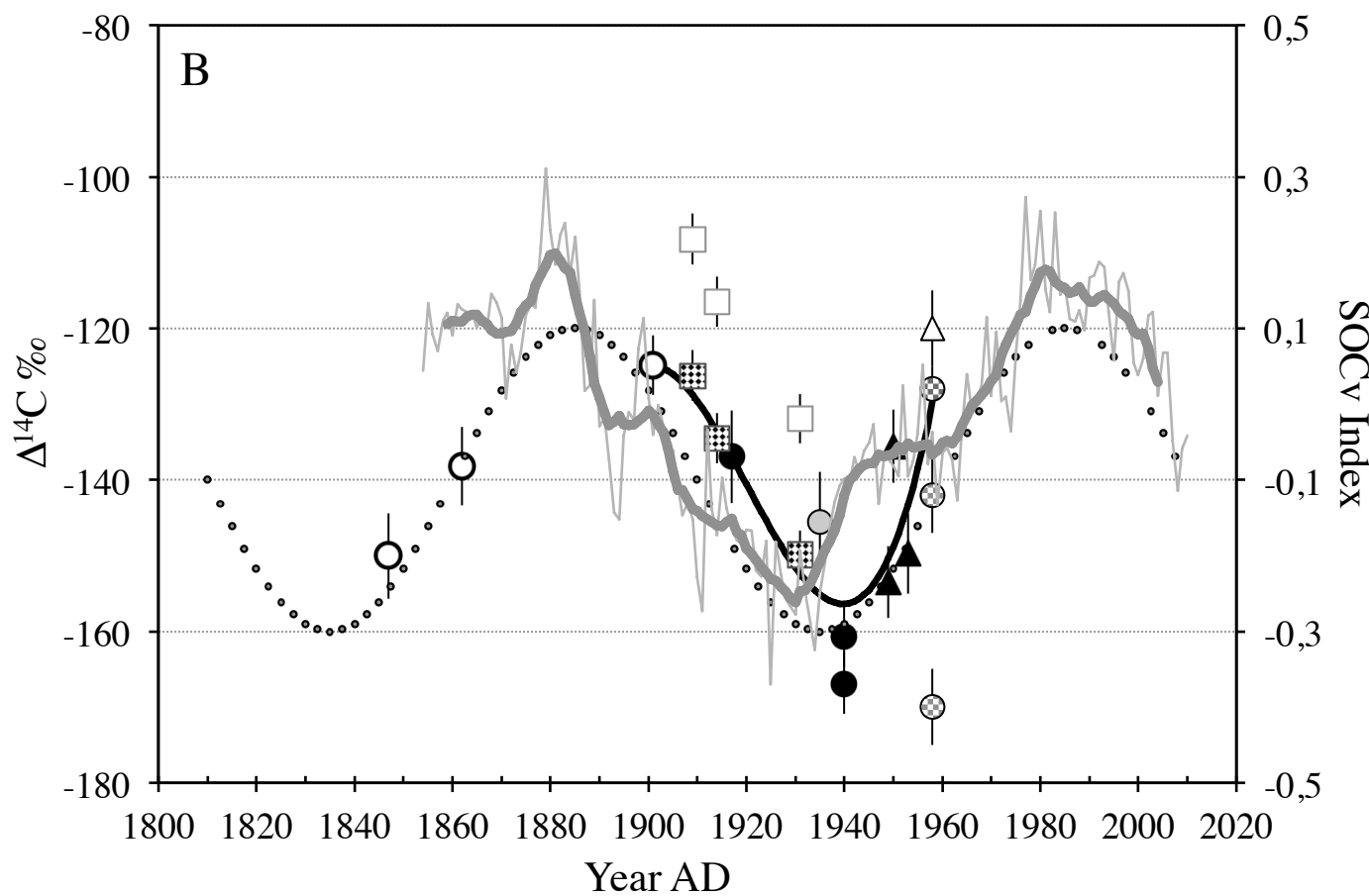
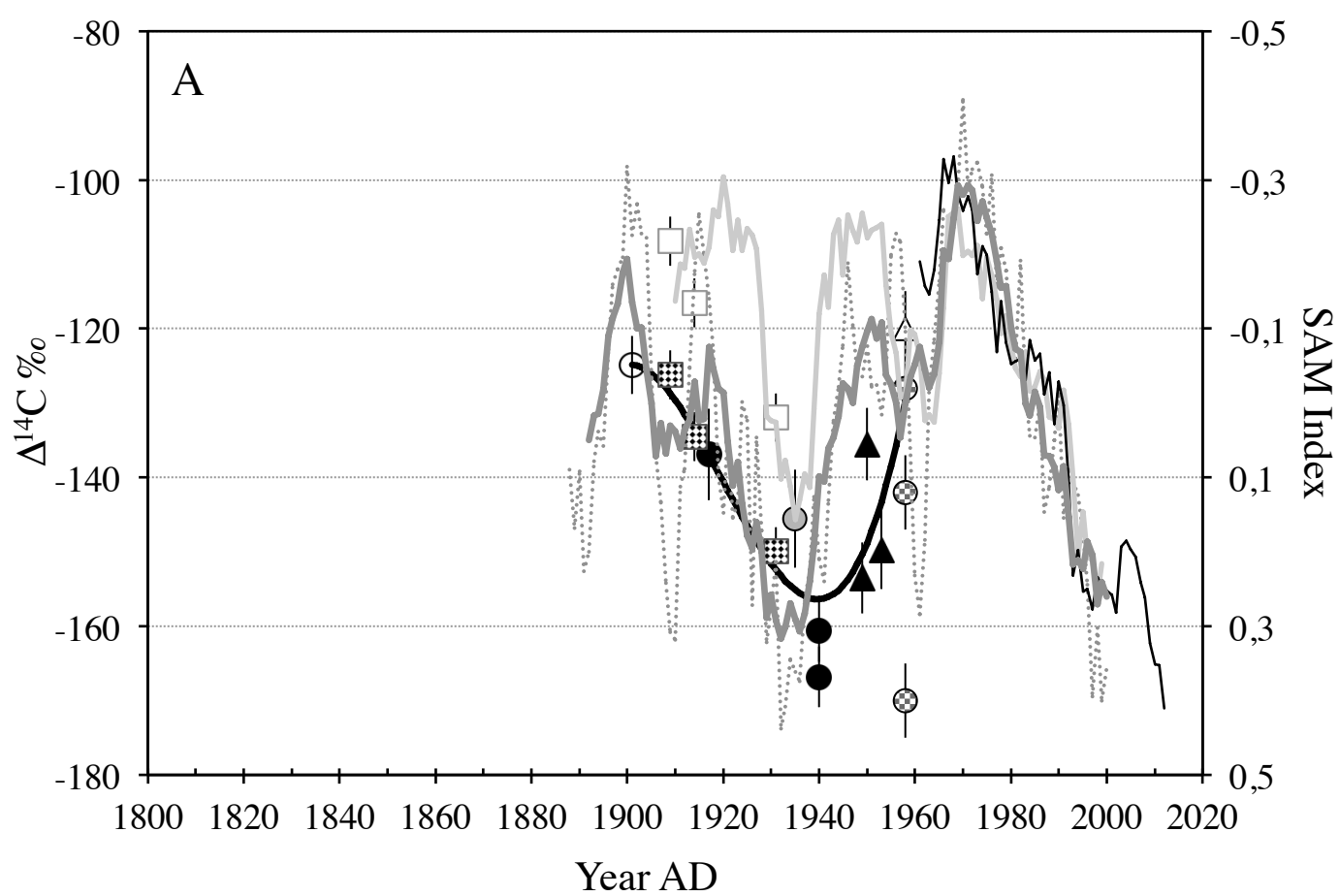


Fig. 4