

February 26, 2018  
Gif-sur-Yvette, France

Dear Alessio Rovere,

Thank you again for reviewing our manuscript and for allowing for an extension to the revision deadline. I was a bit busy due to having moved countries since the manuscript was originally submitted and the extension was much welcomed!

The open discussion format of *Climate of the Past* has allowed us to process some very interesting comments by the reviewers and discussion contributors. The main changes to the manuscript relate to the consideration of bioturbation upon the core material. Following the helpful comments of reviewer Ascough and discussion contributors Dolman et al., we decided to simulate multiple bioturbation scenarios and how they relate to the single foraminifera  $^{14}\text{C}$  data that we have found. In the previous manuscript, we proposed that normally distributed age-depth sediment relationship could explain the bioturbation we see, whereas Dolman et al pointed out that the traditional bioturbation model of Berger and Heath (1968) would predict an exponential age-depth relationship. After carrying out multiple sediment core simulations, and also considering other forms of bioturbation, we have found that the observed PDSM in core T86-10P can best be explained by multiple bioturbation processes, which neither a normally distributed nor exponentially distributed age-depth relationship can fully explain. Specifically, we find that we can reproduce the bioturbation in T86-10P when we simulate much deeper, secondary bioturbation processes documented by (LÖwemark and Grootes, 2004; Löwemark and Werner, 2001; Löwemark and Schäfer, 2003). These secondary processes are not included in traditional bioturbation models (e.g., those by Berger and Heath 1968, Berger and Johnson, 1978; Berger and Killingley, 1982; Guinasso and Schink, 1975; Peng et al., 1979). We have, therefore, created our own bioturbation model which combines the primary bioturbation processes predicted by traditional bioturbation models with the secondary, much deeper *Zoophycos* bioturbation. We find that the combination of these two types of bioturbation can reconcile the single foraminifera  $^{14}\text{C}$  data that we see in core T86-10P. We also felt it would be better to replace the contour plot simulation output in the original submission with a more reader friendly figure which visually shows the age-depth distribution for our single foraminifera simulations.

The changes we outline are slightly more extensive than those we originally outlined in the response to the reviewers, but we feel they are worthwhile as they reconcile the observed single foraminifera  $^{14}\text{C}$  data in core T86-10P by integrating multiple models of bioturbation. We note that our main end product of the manuscript, the dual  $^{14}\text{C}$  and stable isotope analysis, remains unchanged from the original manuscript.

We feel that the *Climate of the Past* open discussion forum has served to improve our manuscript and affirms our choice in submitting to this journal. We'd like to once again warmly thank the reviewers and discussion contributors.

We thank you for considering our manuscript and hope you find the improved manuscript to be suitable for publication in *Climate of the Past*. Appended to this letter is the 'track-changes' version of the manuscript (which MS Word has somewhat exaggerated!).

On behalf of the co-authors,

Kind regards,

Bryan Lougheed

# Moving beyond the age-depth model paradigm in deep sea palaeoclimate archives: dual radiocarbon and stable isotope analysis on single foraminifera.

Bryan C. Lougheed<sup>1,2\*</sup>, Brett Metcalfe<sup>2,3\*</sup>, Ulysses S. Ninnemann<sup>4</sup> and Lukas Wacker<sup>5</sup>

<sup>1</sup>Department of Earth Sciences, Uppsala University, Villavägen 16, 75236 Uppsala, Sweden.  
<sup>2</sup>Laboratoire des Sciences du Climat et de l'Environnement, LSCE/IPSL, CEA-CNRS-UVSQ, Université Paris-Saclay, France.  
<sup>3</sup>Department of Earth Sciences, Faculty of Sciences, Vrije Universiteit Amsterdam, De Boelelaan 1085, 1081HV Amsterdam, the Netherlands.  
<sup>4</sup>Department of Earth Science, University of Bergen, Allégaten 41, 5007 Bergen, Norway.  
<sup>5</sup>Laboratory for Ion Beam Physics, ETH Zürich, Otto-Stern-Weg 5, 8093 Zürich, Switzerland.

\* contributed equally to this study.

Correspondence to: Bryan C. Lougheed (bryan.lougheed@geo.uu.se|lsce.ipsl.fr)

**Abstract.** Late-glacial palaeoclimate reconstructions from deep-sea sediment archives provide valuable insight into past rapid changes in ocean chemistry, ~~but~~. Unfortunately, only a small proportion of the ocean floor with suitably high sediment accumulation rate (SAR) is suitable for such reconstructions using the ~~existing state of the art using the~~ longstanding age-depth model approach. We employ ultra-small radiocarbon (<sup>14</sup>C) dating on single microscopic foraminifera to demonstrate that the longstanding age-depth model method conceals large age uncertainty caused by post-depositional sediment mixing, meaning that existing studies may underestimate total geochronological error. ~~To overcome these problems, we use~~ We find that the age-depth distribution of our <sup>14</sup>C-dated single foraminifera is in good agreement with existing bioturbation models when one takes the possibility of *Zoophycos* burrowing into account. To overcome the problems associated with the age-depth paradigm, we use the first ever, dual <sup>14</sup>C and stable isotope (δ<sup>18</sup>O and δ<sup>13</sup>C) analysis on single microscopic foraminifera to produce a palaeoclimate time series independent of the age-depth paradigm. ~~This new and novel method will~~ This new state-of-the-art essentially decouples single foraminifera from the age-depth paradigm to provide multiple floating, temporal snapshots of ocean chemistry, thus allowing for successful extraction of temporally accurate palaeoclimate data from low SAR deep sea archives. This new method can address large geographical gaps in late-glacial benthic palaeoceanographic reconstructions by opening up vast areas of previously disregarded, low SAR, deep-sea archives, ~~leading to research, which will lead~~ to improved understanding of the global interaction between oceans and climate.

## 1 Introduction

~~The past seven decades in palaeoceanography research have produced a wealth of valuable palaeoclimate data from the calcareous, foraminiferal ooze contained in deep sea sediment archives, greatly improving our understanding of past ocean~~

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chemistry and palaeoclimate (Bond et al., 1993; Emiliani, 1955; Shackleton, 1967). The longstanding geochronological state of the art that has been applied to these sediment archives since the inception of palaeoceanography as a field of study, the *age depth model* method, relies on the geological law of superposition. This law states that sediment age increases progressively with sediment core depth. The method involves first slicing deep sea sediment cores into discrete core depth intervals of 1 cm thickness or greater. Sufficient numbers of suitable foraminifera tests are subsequently picked from select intervals and analysed using mass spectrometry with ages being inferred using  $^{14}\text{C}$  dating and/or orbital tuning (Martinson et al., 1987; Pisias et al., 1984) of  $\delta^{18}\text{O}$  values. Finally, statistical methods (Blaauw and Christen, 2011; Bronk Ramsey, 2008) are used to interpolate ages for all discrete depth intervals of the sediment core.

Post depositional sediment mixing (PDSM) of deep sea archives (due to, e.g., bioturbation) can cause the relationship between sediment core age and sediment core depth to become more complex. This complexity can be masked from researchers because, up until now, successful stable isotope ratio mass spectrometry (IRMS) or  $^{14}\text{C}$  accelerated mass spectrometry (AMS) analysis has required the analysis multi specimen samples containing tens (Metcalfe et al., 2015; Waelbroeck et al., 2005) and hundreds (Hughen et al., 2006) of single foraminifera tests, respectively. IRMS and AMS analyses only report a mean sample value and a machine measurement error, meaning that no information is provided about the true age uncertainty of a discrete depth interval, which is a function of both sediment accumulation rate (SAR) and PDSM. Subsequently, high resolution sampling of core depth does not necessarily translate into high resolution sampling of time. Researchers are aware that the concealment of intra sample age heterogeneity can pose problems for the age depth model method (Bard, 2001; Keigwin and Gagnon, 2015; Löwemark and Grootes, 2004; Löwemark and Werner, 2001; Pisias, 1983; Ruddiman and Glover, 1972), which can potentially lead to incorrect interpretation of temporal climate offsets, or apparent  $^{14}\text{C}$  age offsets between different species and/or sizes of foraminifera that are, in fact, an artefact of PDSM (Löwemark et al., 2008; Löwemark and Grootes, 2004). With these problems in mind, researchers seeking to reconstruct rapid (i.e. sub-millennial and centennial) climate processes have concentrated on sediment archives with a SAR greater than 10 cm/ka (Hodell et al., 2015; Shackleton et al., 2000; Vautravers and Shackleton, 2006), with the assumption that high SAR minimises the effects of PDSM upon age depth models. However, the inability to quantify intra sample age heterogeneity means that this assumption has yet to be rigorously tested. Moreover, the vast majority of the ocean floor exhibits a SAR of less than 10 cm/ka (Fig. 1), meaning that many potentially useful study sites above the calcite compensation depth are essentially rendered unusable by the longstanding geochronological state of the art. Concentrating only on these select parts of the ocean floor with high SAR introduces a geographical bias into our understanding of global ocean processes.

In this study, we utilise the latest developments in ultra small ( $<100\text{ }\mu\text{g CaCO}_3$ ) sample  $^{14}\text{C}$  dating (Synal et al., 2007; Wacker et al., 2013a, 2013b, 2013e). The past seven decades in palaeoceanography research have produced a wealth of valuable palaeoclimate data from the calcareous, foraminiferal ooze contained in deep sea sediment archives, greatly improving our understanding of past ocean chemistry and palaeoclimate (Bond et al., 1993; Emiliani, 1955; Epstein et al., 1951; Shackleton, 1967; Urey, 1947). The longstanding geochronological method that has been applied to these sediment archives since the inception of palaeoceanography as a field of study, the *age-depth model* method, relies on the geological

law of superposition. This law states that sediment age increases progressively with sediment core depth. The age-depth model method, as applied to deep-sea sediment cores, involves first slicing deep sea sediment cores into discrete core depth intervals of 1 cm thickness or greater. Sufficient numbers of suitable foraminifera tests are subsequently picked from select intervals and analysed using mass spectrometry, whereby ages are inferred through  $^{14}\text{C}$  dating and/or orbital tuning (Martinson et al., 1987; Pisias et al., 1984) of  $\delta^{18}\text{O}$  values. Finally, statistical methods (Blaauw and Christen, 2011; Bronk Ramsey, 2008) are used to interpolate ages for all discrete depth intervals of the sediment core.

Post-depositional sediment mixing (PDSM) of deep sea archives (due to, e.g., bioturbation) can cause the relationship between sediment core age and sediment core depth to become more complex, thereby limiting the precise application of the law of superposition. This complexity can be masked from researchers who use the age-depth model method because successful stable isotope ratio mass spectrometry (IRMS) or  $^{14}\text{C}$  accelerator mass spectrometry (AMS) analysis has traditionally required the analysis of multi-specimen samples containing tens (Metcalfe et al., 2015; Waelbroeck et al., 2005) and hundreds (Hughen et al., 2006) of single foraminifera tests, respectively. IRMS and AMS analyses only report a mean sample value and a machine measurement error, meaning that no information is provided about the true heterogeneity of the sample, which is chiefly a function of sediment accumulation rate (SAR) and PDSM. As such, the true age uncertainty within a sediment core can remain hidden from the researcher. Furthermore, AMS analysis reports a sample's mean  $^{14}\text{C}$  activity, which is not the same as a sample's mean  $^{14}\text{C}$  age, which creates further complications for highly heterogeneous samples (as alluded to by reviewer Ascough [2017]).

Considering the aforementioned complications associated with the age-depth model method, high resolution sampling of core depth does not necessarily translate into high resolution sampling of time. Researchers are aware that the concealment of intra-sample age heterogeneity can pose problems for the age-depth model method (Bard, 2001; Keigwin and Gagnon, 2015; Löwemark and Grootes, 2004; Löwemark and Werner, 2001; Pisias, 1983; Ruddiman and Glover, 1972), which can potentially lead to incorrect interpretation of temporal climate offsets, or apparent  $^{14}\text{C}$  age offsets between different species and/or sizes of foraminifera that are, in fact, an artefact of PDSM (Berger, 1977; Löwemark et al., 2008; Löwemark and Grootes, 2004; Peng and Broecker, 1984). With these problems in mind, researchers seeking to reconstruct rapid (i.e. sub-millennial and centennial) climate processes have generally concentrated on sediment archives with a SAR greater than 10 cm/ka (Hodell et al., 2015; Shackleton et al., 2000; Vautravers and Shackleton, 2006), with the assumption that high SAR minimises the effects of PDSM upon age-depth models. However, the inability to directly quantify intra-sample age heterogeneity means that this assumption has yet to be rigorously tested. Furthermore, the vast majority of the ocean floor exhibits a SAR of less than 10 cm/ka (Fig. 1), meaning that many potentially useful study sites above the calcite compensation depth are essentially rendered unusable by the longstanding age-depth approach. Concentrating only on the select parts of the ocean floor with high SAR introduces a geographical bias into our understanding of global ocean processes.

In this study, we utilise the latest developments in ultra-small ( $<100\text{ }\mu\text{g CaCO}_3$ ) sample  $^{14}\text{C}$  dating (Synal et al., 2007; Wacker et al., 2013a, 2013b, 2013c) to reduce sample size to a single benthic foraminifer specimen (*Cibicidoides*

wuellerstorfi), thereby allowing us to directly quantify intra-sample age heterogeneity and statistically analyse PDSM in the case of the low SAR (~1-2 cm/ka) sediment core T86-10P from the Azores region of the North Atlantic (Fig. 1). We discuss the consequences of our results for existing studies and provide a framework suggestions for adding realistic geochronological errors to existing deep sea palaeoclimate records that have applied the longstanding age-depth model method- using multi-specimen samples. Furthermore, we analyse both <sup>14</sup>C and stable isotopes on select single foraminifer tests specimens of sufficient mass, demonstrating the possibility feasibility to construct palaeoclimate time series that are completely independent of the age-depth paradigm and its associated problems, thereby opening up vast, low SAR regions of the ocean to late glacial palaeoceanography research, which will lead to a more integrated global picture of ocean ventilation and water mass reorganisation processes during the last deglaciation, a period of rapid climate change.

## 2 Method

### 2.1 Sediment core retrievals selection and subsampling

Sediment core T86-10P (Fig. 1) was retrieved from the North Atlantic (37° 8.13' N, 29° 59.15' W, 2610 mbsl) by the vessel R/V Tyro as part of the APNAP-I project. We chose core T86-10P for this study because preliminary oxygen isotope measurements on planktonic foraminifera indicated the possible a poor multi-specimen glacial-interglacial δ<sup>18</sup>O stratigraphy, typical of a low SAR sediment core (Metcalf, 2013). Also, the nearby presence of significant PDSM, a very high SAR record at a similar water depth (Repschläger et al., 2015) provides an ideal local 'reference' stratigraphy for direct comparison. In other words, core T86-10P is an ideal sediment core with which to test the ability of dual <sup>14</sup>C and stable isotope analysis on single foraminifera to successfully extract temporally accurate palaeoclimate data from a very low SAR archive.

185 single specimen benthic foraminifera tests (*Cibicidoides wuellerstorfi*)wuellerstorfi, the same species used in Repschläger et al. [2015], ranging between 25 and 500 µg in mass were retrieved from the wet sieved and deionised water washed >250 µm fraction of 1 cm slices of the sediment core material. 47 discrete 1 cm intervals were picked for foraminifera, with an average of 3.9 specimens per discrete interval being picked (min 1 specimen, max 10 specimens). Care was taken to pick whole tests that had not been subjected to exhibited neither physical damage nor visible dissolution. Of these 185 tests, 100 were measured for stable isotopes only (δ<sup>18</sup>O and δ<sup>13</sup>C) and 49 were measured for <sup>14</sup>C only, while 36 tests of sufficient mass were successfully analysed for both <sup>14</sup>C and stable isotopes. These 36 tests were cut using a scalpel (future work may use more efficient gaseous splitting) according to an approximate 80/20 ratio, with the larger fraction being reserved for <sup>14</sup>C AMS analysis and the smaller fraction being reserved for stable isotope IRMS analysis. All data are available in spreadsheet format in Table S1.

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## 2.2 $^{14}\text{C}$ analysis

AMS analysis was carried out at the Laboratory for Ion Beam Physics at ETH Zürich using a permanent magnet equipped Mini Carbon Dating System (MICADAS) AMS (Synal et al., 2007) (Synal et al., 2007) with Helium stripping system, coupled to an Ionplus carbonate handling system (Wacker et al., 2013a) (Wacker et al., 2013a). The MICADAS setup allows for direct  $^{14}\text{C}$  measurement, using a gas ion source (Wacker et al., 2013b) (Wacker et al., 2013b), of gaseous  $\text{CO}_2$  liberated from  $\text{CaCO}_3$  samples by acidification with phosphoric acid – i.e. no graphitisation step is necessary. The exclusion of the graphitisation step allows for the required sample mass to be reduced down to 100  $\mu\text{g}$  of  $\text{CaCO}_3$  (12  $\mu\text{g}$  C) and less (Lougheed et al., 2012; Wacker et al., 2013c) (Lougheed et al., 2012; Wacker et al., 2013c), enabling sample size to be reduced to one specimen in the case of a suitable foraminifera species and specimen. Procedural single specimen foraminifera blank samples (assumed Eemian age) from core T86-10P indicated an average mean blank value of 0.0115  $\text{F}^{14}\text{C}$  (n=10). Procedural IAEA-C1 standard blank material of similar mass as the single foraminifera tests yielded an average blank value of 0.0100  $\text{F}^{14}\text{C}$  (n=10) a mean blank value of 0.0100  $\text{F}^{14}\text{C}$  (n=10). No relationship was found between blank mass and blank value. Representative blanks (i.e. those not affected by flux jumps, etc.) were pooled and a specific blank correction of  $0.0095 \pm 0.002 \text{ F}^{14}\text{C}$  was applied to most of the samples using the BATS software (Wacker et al., 2010). In the case of a small group of remaining samples, which came from a second run (those with lab code prefix ETH-74), a lower blank correction ( $\text{F}^{14}\text{C} = 0.0033 \pm 0.001$ ) was applied. Reported  $^{14}\text{C}$  ages (Table S1) are rounded following standard conventions (Stuiver and Polach, 1977). Our small single specimen tests are slightly more susceptible to the influence of secondary carbonate phases, because pre-treatment was often not possible on such small samples. To control for this effect, Research is ongoing regarding achieving a superior blank value for such small samples, which would allow us to improve precision and further push back the age limit of single foraminifera  $^{14}\text{C}$  analysis. The blank values we have achieved in this study, however, are sufficient for quantifying PDSM and reconstructing deglacial processes (i.e. the past 20 ka) in the low SAR core T86-10P. Our  $^{14}\text{C}$  measurement precision is one order of magnitude smaller than the reconstructed PDSM, while the general geochronological uncertainties associated with  $^{14}\text{C}$  age calibration of marine samples (i.e. reservoir age and calibration curve uncertainties) are generally greater than our measurement uncertainty. It could be argued that high precision  $^{14}\text{C}$  analysis within the marine realm is not strictly necessary. Moreover, especially when studying highly heterogeneous material, sacrificing precision to reduce sample size to one specimen translates to more accurate results than when one carries out high precision measurements on multi-specimen samples – in the latter case one loses all information regarding the true age precision, i.e. the intra-sample age heterogeneity. We carried out experiments to investigate the possible presence of secondary carbonate phases in our samples; select single specimens (n=5) of sufficient size were  $^{14}\text{C}$  analysed for both an initial phosphoric acid leach fraction and the residual foraminifer fraction. Of these five specimens, three specimens yielded leach and residual  $^{14}\text{C}$  ages that were not significantly different within  $1\sigma$ , and a fourth specimen was not significantly different within  $2\sigma$  (the fifth was specimen was not significantly different within  $2.1\sigma$ ).

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165 Benthic *C. wuellerstorfi*  $^{14}\text{C}$  ages were calibrated using *MatCal 2.2* (~~Lougheed and Obrochta, 2016~~)(Lougheed and Obrochta, 2016), employing the *Marine13* (~~Reimer and et al., 2013~~)(Reimer et al., 2013),  $^{14}\text{C}$  calibration curve and an appropriate marine reservoir age ( $\Delta\text{R}=35\pm290$   $^{14}\text{C}$  yr), the latter of which was calculated as follows: Previous studies in this region using planktonic foraminifera have employed the standard marine calibration curve (i.e.  $\Delta\text{R}=0\pm0$ ), but the possibility of spatiotemporally dynamic  $\Delta\text{R}$  for the Azores region has been alluded to previously (Schwab et al., 2012; Waelbroeck et al., 2001). We are aware of the potential uncertainties associated with  $\Delta\text{R}$ , so we employ a planktonic  $\Delta\text{R}$  with a large uncertainty:  $0\pm200$   $^{14}\text{C}$  yr. Seeing as we carried out our investigation upon benthic foraminifera, we must additionally take into account the possibility that benthic  $\Delta\text{R}$  may be different from that of the ocean's surface mixed layer- for which *Marine13* was developed. Using available data from a nearby sediment core-~~archive~~, from the Azores region (MD08-3180) (Sarnthein et al., 2015), we analyse  $^{14}\text{C}$  determinations from a late-glacial sequence of co-occurring benthic and planktonic foraminifera (S1 and Fig. S1). We find that the long-term (7 ka) average  $^{14}\text{C}$  age difference between the planktonic and benthic foraminifera is  $35\pm210$   $^{14}\text{C}$  yr, suggesting that there is only a small absolute difference between benthic and planktonic  $^{14}\text{C}$  ages in this region, but with considerable variation. We arrive at our final benthic  $\Delta\text{R}$  by correcting our planktonic  $\Delta\text{R}$  ( $0\pm200$   $^{14}\text{C}$  yr) for the benthic offset ( $35\pm210$   $^{14}\text{C}$  yr) with error propagation, resulting in a final  $\Delta\text{R}$  of  $35\pm290$   $^{14}\text{C}$  yr.

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### 2.3 Stable isotope analysis

180 IRMS analysis on the smaller foraminifera test fractions from the cut tests (as well as some whole tests) was carried out at the stable isotope laboratory of the Department of Earth Science, University of Bergen, using a *Kiel IV* carbonate device coupled to a *Thermo MAT-253* dual inlet IRMS. The use of a dual-inlet ~~IMRS~~IRMS, as opposed to a continuous flow ~~mass spectrometer~~IRMS, leads to a reduced size difference between sample and standard gas, combined with a continuous switching between standard and sample gas, which enables a higher analytical precision- for small samples. Procedural standard samples (Carrara marble powder) of representative mass indicated an external precision ( $1\sigma$ ) better than 0.10 ‰ and 0.05 ‰ for  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ , respectively. Additional whole tests were analysed using a *GasBench II* preparation device coupled to a continuous flow *Thermo Delta+ Plus* mass spectrometer at the Department of Earth Sciences, Vrije Universiteit Amsterdam. For these measurements, the external precision ( $1\sigma$ ) of international standards was better than 0.12 ‰ for both  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  (~~Feldmeijer et al., 2015; Metcalfe et al., 2015~~)(Feldmeijer et al., 2015; Metcalfe et al., 2015). All IRMS measurements are reported in per mil (‰) against the Vienna Pee Dee Belemnite (V-PDB) scale.

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## 3 Results and Discussion

### 3.1 Age uncertainties concealed by the ~~current state of the art~~longstanding geochronological method

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190  $^{14}\text{C}$  analysis carried out on 85 single specimen foraminifera tests for core T86-10P (Fig. 2A, Table S1) indicate the presence of significant PDSM, with the average standard deviation for all discrete 1 cm core depth intervals containing three

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or more  $^{14}\text{C}$  dates being 46704667  $^{14}\text{C}$  yr. We show that such significant PDSM can be concealed by ~~the current~~longstanding geochronological ~~state of the art, by imitating the longstanding methods. Specifically, we imitate the age-depth model approach involving multi-specimen samples. To achieve this, we average by averaging all uncalibrated single specimen  $^{14}\text{C}$  data from discrete depths with three or more measured single specimens into pseudo multi-specimen  $^{14}\text{C}$  dates with an uncertainty of 60  $^{14}\text{C}$  yr (a typical AMS machine error for larger samples). We subsequently calibrate these pseudo multi-specimen dates and produce a Bayesian age-depth model for core T86-10P (Fig. 2B) using the Bacon software (Blaauw and Christen, 2011).~~2B) using the Bacon software (Blaauw and Christen, 2011). This age model displays an apparent average SAR of 2.2  $\pm$  0.9 cm/ka, with higher apparent SAR in the uppermost 10 cm of the core than the lower parts of the core, which is consistent with typically observed sediment mixed layer depths (e.g. Trauth et al., 1997). Considering the large intra-sample heterogeneity present in core T86-10P, our pseudo multi-specimen Bayesian age-depth model exhibits unrealistically well-constrained confidence intervals, thus concealing the true age-depth variation present within core T86-10P. Additionally, the Bayesian age-depth modelling routine excludes a number of pseudo multi-specimen dates as outliers. Rather than being regarded as outliers, we propose that downcore multi-specimen  $^{14}\text{C}$  dates in non-sequential temporal order may actually serve as useful indicators for the presence of significant PDSM throughout an entire sediment sequence, rather than being regarded as outliers related to isolated events, as is often done in the literature.

While core T86-10P may not be representative of all sediment cores, the fact that very large intra-sample age heterogeneity can be concealed by ~~the longstanding geochronological state of the art~~methods, has significant implications for existing studies relying on the age-depth model method. We note that our pseudo multi-specimen  $^{14}\text{C}$  dates were assembled using data from an average of four foraminifera tests each. Typically, multi-specimen samples containing many tens to hundreds of foraminifera tests are used for  $^{14}\text{C}$  dating. Were such large sample sizes to be applied to T86-10P, it is possible that no age-depth outliers would be ~~present~~produced, and that all information about intra-sample heterogeneity would be lost, thus concealing the full temporal uncertainty from the researcher. Such an affect was seen in one of the earliest studies using ultra-small  $^{14}\text{C}$  dating of foraminifera samples (Lougheed et al., 2012), whereby downcore age-depth reversals were found for a sequence of multi-specimen samples with <500  $\mu\text{g}$  mass, whereas a sequence of multi-specimen samples with a greater mass did not exhibit such a behaviour.

We carried out simulations to investigate the influence of  $^{14}\text{C}$  sample size upon the concealment of age-depth outliers by using multiple simulated synthetic sediment core scenarios whereby exaggerated single foraminifera PDSM is generated using Gaussian noise (S2 and Fig. S2). These preliminary simulations suggest that when sample size is five to ten specimens or more, no age-depth outliers are present in a simulated sediment core with intense PDSM. ~~Considering that the longstanding state of the art in  $^{14}\text{C}$  analysis requires up to hundreds of foraminifera specimens (e.g., Hughen et al., 2006), it is possible that the full extent of PDSM has been hidden by existing methods at certain study locations.~~

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225 **3.2 Quantifying intra-core post depositional movement of Core T86-10P in the context of existing bioturbation understanding**

Seeing as it has not been previously possible to  $^{14}\text{C}$  date single foraminifera, past research of PDSM has focussed on quantifying bioturbation processes by constructing theoretical models. One of the earliest models for bioturbation in deep sea sediment cores by Berger and Heath (1968) assumes that the uppermost layer of sediment core is uniformly to a certain mixed layer depth (typically 10 cm) throughout the sedimentation history of the core. This general model of bioturbation has formed the basis for subsequent modelling investigations into vertical PDSM of sediment particles of different age (e.g., Berger and Johnson, 1978; Berger and Killingley, 1982; Guinasso and Schink, 1975; Peng et al., 1979). A general feature of these traditional bioturbation models is that the continuous application of a uniformly mixed upper layer of sediment throughout the entire sedimentation history of given core archive will result in an exponential probability density function (PDF) for age at any given core depth. Such an exponential PDF will exhibit a maximum probability for younger ages, with a long tail towards older ages.

———— We can construct an ideal superposition ranking of the single foraminifera To determine if the single foraminifera  $^{14}\text{C}$  data we retrieved from core T86-10P can be approximated using the aforementioned bioturbation models, we have carried out a single foraminifera sedimentation simulation using a uniform mixed layer depth of 10 cm and constant SAR of 1.4 cm/ka. The SAR applied is on the low end of the SAR estimated by the *Bacon* age-depth model ( $2.2 \pm 0.9$  cm/ka), because we have considered that much of the *Bacon* age model also includes the interval of the sediment core within the active mixed layer depth. We consider, therefore, that 1.4 cm/ka represents a good estimation the core SAR outside the mixed layer depth (i.e. the SAR corresponding to the historical layers). Our simulation is carried out in sedimentation intervals of 0.1 cm, with 400 new foraminifera being added per interval and assigned an age according to the SAR. At each sedimentation interval, the upper 10 cm of the sediment is uniformly mixed using random noise. The results of the simulation are presented in Figs. 3A and 3B, and superimposed upon them are the calibrated ages for all single foraminifera that we have  $^{14}\text{C}$  dated. The simulation would seem to suggest that the population of single foraminifera we have  $^{14}\text{C}$  dated in our study cannot be approximated using a sedimentation simulation that uses only a uniformly mixed layer depth, i.e. the method used by traditional bioturbation models. While we apply a constant SAR, using a dynamic SAR in our simulation would not resolve this disagreement, because it would simply shift the median age towards certain  $^{14}\text{C}$ -dated single foraminifera, but away from others (Fig. 3B). It is apparent that the age-depth distribution of our single foraminifera is not compatible with the exponential age PDF predicted by traditional bioturbation models. Most notably, such bioturbation models cannot explain the downward movement of young foraminifera to great depth in T86-10P, as in the case of the specimen at 41-42 cm depth (7.9 cal ka) or the one at 21-22 cm depth (1.2 cal ka) (Fig. 3B).

255 To further analyse the age-depth distribution of the  $^{14}\text{C}$  dated single foraminifera in T86-10P, we construct an ideal superposition ranking of the single foraminifera (i.e. in the case of no PDSM) from core T86-10P by ranking them by median calibrated  $^{14}\text{C}$  age. We can also rank the foraminifera by sediment core depth, i.e. by the 1 cm depth interval they were actually retrieved from. 'Depth ranking' is not the same as 'depth', but nonetheless provides an interesting way to

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visualise the movement of single foraminifera. By comparing the ~~two rankings~~ age ranking and depth ranking, we can visualise the post-depositional upcore and downcore movement of the single foraminifer tests (Fig. 3A4A). Analysis of the post-depositional ranking change for ~~all the~~ foraminifera indicates that while the ranking change appears normally distributed, ~~but it~~ just fails a ~~Kolmogorov-Smirnov (K-S)~~ statistical test for normal distribution (Fig. 3B). However, the youngest and oldest ~~4B~~. It would appear, therefore, that the age distribution of the single foraminifera within our population are biased to show minimal ranking change, as they are at the temporal edge of our population and that we do not have the ability to sample into infinity. To overcome this 'edge-effect', we subsequently exclude the 20 youngest and 20 oldest foraminifera, after which the ranking change for the middle of our population can <sup>14</sup>C dated cannot be described as normally distributed fully approximated using the K-S test procedure (Fig. 3C). No correlation was found between foraminifera size and ranking change (S3 and Fig. S3). The ranking change and K-S test procedure was repeated  $10^6$  times (each time analysing the computed middle of the population) using probability weighted random sampling of the calibrated age PDF of each foraminifera, and the ranking change was found to be normally distributed for 99.98% of the  $10^6$  runs a normal distribution, nor by the exponential distribution suggested by traditional bioturbation models.

The indication that the effect of PDSM upon single foraminifera age-depth distribution in sediment core T86-10P can be approximated using a normal distribution allows us to use Gaussian noise to explore theorised intra-sample age heterogeneity for multiple synthetic sediment core scenarios. We simulate the age heterogeneity (i.e. the  $1\sigma$  age value) of  $10^5$  synthetic foraminifera within a 1 cm sediment core slice in the case of  $10^4$  sediment core scenarios involving  $10^3$  SAR scenarios and  $10^3$  PDSM intensity scenarios (Fig. 4). Using an estimated SAR for T86-10P of  $2.2 \pm 0.9$  cm/ka (Fig. 2B), and knowing that the average intra-sample age heterogeneity ( $1\sigma$  age value) for discrete 1 cm depths in core T86-10P is 4670 <sup>14</sup>C yr, we can plot the approximate interval of core T86-10P in Fig. 4. The resulting interval suggests that the post depositional movement of single foraminifera is at least 6 cm ( $1\sigma$ ).

It may be possible that secondary bioturbation processes are contributing to the observed PDSM in T86-10P. The burrows of an unidentified organism, referred to as ichnofacies *Zoophycos* burrows, has been shown to penetrate much farther down into the sediment than the uniform mixed layer depth (LÖwemark and Werner, 2001; Wetzel and Werner, 1980). These burrows are often invisible to the naked eye, with X-ray radiographs being necessary for identification. Such secondary bioturbation effects are not considered by traditional bioturbation models, a fact that researchers have previously noted (LÖwemark and Grootes, 2004; LÖwemark and Werner, 2001). The practical effect of *Zoophycos* secondary bioturbation upon a given discrete depth interval would be to introduce a population of significantly younger sediment from above (LÖwemark and Schäfer, 2003), thus altering the age distribution of that discrete depth interval. The potential presence of *Zoophycos* may, therefore, offer an explanation for the apparent disagreement between the single foraminifera <sup>14</sup>C age-depth relationship for core T86-10P and that predicted by our sedimentation simulation when only the traditional model of bioturbation is applied (Fig. 3B). To further investigate this possibility, we carry out a new single foraminifera sedimentation simulation in Figs. 3C and 3D. This simulation is forced using the same SAR (1.4 cm/ka) and mixing layer depth (10 cm) parameters as the simulation previously described in Figs. 3A and 3B, but with the addition of a post-simulation bioturbation by *Zoophycos*.

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Specifically, we take 10% of the total single foraminifera population from the entire core and add an additional randomly selected depth between 0 and 50 cm to their depth values. This process essentially serves to simulate 10% of the core sediment being bioturbated downwards by *Zoophycos*. The addition of the *Zoophycos* procedure to our simulation produces a simulated foraminifera age-depth relationship that can reconcile the presence of the young  $^{14}\text{C}$ -dated foraminifera that we find at depth in core T86-10P (Fig. 3C and 3D).

### 3.3 Consequences for ~~the existing studies using longstanding geochronological state-of-the-art methods~~

The fact that significant PDSM can essentially be concealed by the current state of the art has significant consequences for recent studies that use age depth reconstructions from deep sea sediment archives to reconstruct rapid changes in palaeoclimate. Many such studies (Barker et al., 2015; Caley et al., 2012; Simon et al., 2016) use tuning to the LR04 (Lisiecki and Raymo, 2005) benthic stack to produce an age depth chronology. Were the LR04 stack to display similar PDSM uncertainty as T86-10P, one could expect an intra-sample heterogeneity ( $1\sigma$  age value for a 1 cm slice) of between 1500-2000 years (Fig. 4), in addition to any uncertainties related to the tuning process itself, which may be in the order of multiple millennia (Blaauw, 2012; Martinson et al., 1987; Pisias et al., 1984).

For some continental margin sites, such as those from the Iberian Margin, SAR is very high (20-30 cm/ka) and such study sites have been used for centennial resolution age depth climate reconstructions (i.e.  $\pm 50$  years precision), with the assumption that high SAR essentially minimises the effect of bioturbation upon age depth reconstructions (Hodell et al., 2015; Shackleton et al., 2000; Vautravers and Shackleton, 2006). Other studies suggest that such high SAR can be best used for millennial resolution (i.e. not centennial) (Bard, 2001). Our analysis suggests that for such sites (e.g., U1385, one of the “Shackleton sites” (Hodell et al., 2015)), it is only possible to detect centennial resolution from 1 cm sediment core slices when PDSM intensity is almost negligible, i.e. when the  $1\sigma$  value for foraminifera redeposition depth is less than 1 cm (Fig. 4), assuming that one does not use sediment slices thicker than 1 cm. Were PDSM at Iberian margin sites to be as intense as T86-10P, then 1cm intra sample heterogeneity could be between 400 and 800 years ( $1\sigma$ ).

The concealment of PDSM by longstanding geochronological methods presents significant consequences for existing studies that use stratigraphic and geochronological data sourced from deep sea sediment archives to reconstruct rapid changes in palaeoclimate. Many recent such studies (e.g., Barker et al., 2015; Caley et al., 2012; Simon et al., 2016) use tuning to the LR04 (Lisiecki and Raymo, 2005) benthic stack to produce an age-depth chronology. We have rerun our single foraminifera sedimentation simulation using the average SAR of LR04 (3.9 cm/ka) and found that one could expect a relative 68.2% age range of -1330 to 2950 yr for discrete 1 cm depth intervals. When tuning to LR04, one must also consider any uncertainties related to the tuning process itself, which may be on the order of multiple millennia (Blaauw, 2012; Martinson et al., 1987; Pisias et al., 1984).

For some continental margin sites, such as those from the Iberian Margin, SAR is very high (20-30 cm/ka) and such study sites have been used for centennial resolution age-depth climate reconstructions (i.e.  $\pm 50$  years precision), with the assumption that high SAR essentially minimises the effect of bioturbation upon age-depth reconstructions (Hodell et al.,

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2015; Shackleton et al., 2000; Vautravers and Shackleton, 2006). On the other hand, Bard (2001) suggests that such high SAR can at best be used only for millennial resolution (i.e. not centennial). We have rerun our single foraminifera sedimentation simulation using a high SAR (20 cm/ka) typical of Iberian Margin sites (Figs. 3E and 3F). These simulations suggest a relative 68.2% age range of -260 to 570 yr for a discrete 1 cm depth interval.

Specifically in the case of  $^{14}\text{C}$  dating of multi-specimen samples, it is of great important to consider the heterogeneity of the age distribution of a discrete sediment interval. Radiocarbon laboratory results are based on the mean  $^{14}\text{C}$  activity of a sample, to which laboratories apply the Libby half-life in order to report a radiocarbon age in  $^{14}\text{C}$  years (hereafter, “AMS age”).  $^{14}\text{C}$  is a radioactive isotope and its activity relationship with time is exponential, with the consequence that a highly heterogeneous multi-specimen sample may produce an AMS age that is significantly offset from the actual mean  $^{14}\text{C}$  age of the sample. For all three sedimentation scenarios in Figs. 3A, 3C and 3E, we have calculated the relative mean age, median age and AMS age for discrete depth intervals in all three simulation scenarios, allowing us to demonstrate the behaviour of the potential AMS age bias caused by heterogeneous age distributions. Comparing the simulations in Fig. 3A and 3C, it is apparent that the addition of only a relatively small amount of younger material with an exponentially higher  $^{14}\text{C}$  activity can serve to significantly shift the AMS age towards a much younger value. Researchers carrying out both  $^{14}\text{C}$  AMS and stable isotope ( $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ ) analysis on the same sediment core should, therefore, be aware that the AMS age is potentially skewed by an activity bias, while stable isotopes are not. It is paramount, therefore, to consider the effect of discrete depth interval age distribution when carrying out palaeoclimate reconstructions. It is furthermore possible that the discrete depth age distribution of a particular foraminifera species can change throughout the history of a sediment archive as a result of temporal changes in SAR, species abundance, mixed layer depth, *Zoophycos* intensity, etc.

It must be stressed that core T86-10P represents a single sediment archive location and may not be wholly representative for all locations. ~~However,~~ Moreover, our study is based on the analysis of *C. wuellerstorfi* within the >250  $\mu\text{m}$  fraction, whereas foraminifera in smaller fractions may be differently affected by PDSM (Wheatcroft, 1992). An exact quantification of the intra-sample age heterogeneity at other locations is essentially unknown because it can be concealed by the longstanding state-of-the-art geochronological methods. Furthermore, the intra-sample age heterogeneity for less consolidated sediment within actively bioturbated younger sediment sequences may differ from the intra-sample age heterogeneity for older, more consolidated sediment. We propose, therefore, that the ultra-small sample  $^{14}\text{C}$  methods and statistical analysis we outline in this study can be used, in combination with modelling techniques, to definitively help quantify intra-sample age heterogeneity for various sediment sequences at other study locations (including those in the LR04 benthic stack), thus allowing for the application of a suitable downcore geochronological uncertainty. Such an approach will ultimately lead to a better temporal integration of deep sea sediment archives within the global palaeoclimate record.

3.4 Bypassing the age-depth model paradigm?

~~A~~We show that the limitations of the age-depth model paradigm essentially preclude the extraction of temporally useful deglacial benthic ventilation data from very low SAR archives such as core T86-10P. The traditional, discrete depth average

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(multi-specimen) downcore stable isotope stratigraphy for core T86-10P (Figs. 5A and 5B) shows many spurious, large excursions in  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ , indicating that it would not be possible to use the longstanding state of the art (i.e. the age-depth model method) to retrieve temporally useful information about palaeoclimate or benthic ocean ventilation from the low SAR core T86-10P. The underlying cause for these large excursions is revealed by single specimen foraminifer  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  data (Figs. 5A and 5B), which show a large spread in values, an artefact of PDSM of the core material due to, e.g., bioturbation. This spread in values is significantly larger than the machine error associated with IRMS analysis (typically 0.1‰), meaning that it would be masked/concealed by multi-specimen sample IRMS analysis. Dual measurements of both  $^{14}\text{C}$  and stable isotopes ( $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ ) on the same single specimen foraminifer in core T86-10P can contribute palaeoclimate information that is independent of the geological law of superposition, thereby decoupling an individual foraminifer from the sediment archive to provide a floating, temporal snapshot of ocean chemistry. Multiple such snapshots can facilitate a time history of ocean chemistry that is wholly insensitive to sediment core PDSM and associated issues involving multi-specimen samples within the age-depth paradigm, such as spurious age artefacts between foraminifera with different species/morphologies/preservation conditions (Löwemark et al., 2008; Löwemark and Grootes, 2004). Dual measurements of both  $^{14}\text{C}$  and stable isotopes ( $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ ) on the same single specimen foraminifer in core T86-10P can decouple an individual foraminifer from the sediment archive to provide a floating, temporal snapshot of ocean chemistry that is independent of the geological law of superposition, thereby contributing palaeoclimate information wholly independent of the age-depth paradigm. Multiple such snapshots can facilitate a time history of ocean chemistry that is completely insensitive to PDSM induced issues involving multi-specimen samples within the age-depth paradigm, such as spurious age artefacts between foraminifera of different species, abundance changes, SAR changes, morphologies, dissolution/preservation conditions, particle size dependent mixing, etc. (Berger, 1977; Löwemark et al., 2008; Löwemark and Grootes, 2004; Peng and Broecker, 1984; Wheatcroft, 1992). Due to the combined measurement size requirements of both AMS and IRMS, our dual  $^{14}\text{C}$  and stable isotope measurements on single *C. wuellerstorfi* specimens were limited to those of sufficient mass ( $>100\text{ }\mu\text{g CaCO}_3$ ), which are generally less abundant during the coldest stadial conditions, such as the last glacial maximum (LGM); a problem that also affects studies using traditional, multi-specimen reconstructions (e.g., Shackleton et al., 2000). Nevertheless, we (e.g., Shackleton et al., 2000). We were able to produce successful dual  $^{14}\text{C}$  and stable isotope measurements for a sufficient number of foraminifera, revealing a *C. wuellerstorfi* benthic deglaciation signal for core T86-10P. This benthic deglaciation signal is in good agreement with existing *C. wuellerstorfi* data from a previous study using a nearby (140 km proximity) high SAR ( $\sim 20\text{ cm/ka}$ ) record (Repschläger et al., 2015) (Figs. 5C and 5D). Specifically, we find good temporal agreement with the absolute values for  $\delta^{18}\text{O}$ , indicating a valid benthic deglaciation signal. We also find good temporal agreement with a sharp peak in  $\delta^{13}\text{C}$  values that has previously been interpreted as a local increase in Eastern North Atlantic Deep Water (ENADW) linked to the onset of the Holocene (Repschläger et al., 2015). These Our results demonstrate that it is possible to use our dual  $^{14}\text{C}$  and stable isotope method on single foraminifera to extract temporally accurate late glacial/deglaial, benthic

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palaeoceanographic data from a very low SAR site, with success comparable success when compared to data extracted from a very high SAR site using the existing state of the art where traditional methods were used.

#### 4 Conclusion

Analysis of  $^{14}\text{C}$  on single foraminifera opens up new possibilities for quantifying the total geochronological error in existing studies due to PDSM, which. These errors may have been previously underestimated overlooked due to inherent limitations associated with the longstanding geochronological state-of-the-art using method based on multi-specimen species within an the age-depth paradigm. Using the methods outlined in this study, it is possible to quantify the age-depth geochronological uncertainty for existing late-glacial palaeoceanographic records, thus placing them as well as to consider the possibility of AMS age biases associated with very heterogeneous multi-specimen samples. Full consideration of uncertainties will help to place existing palaeoceanographic records within an accurate geochronological framework. Subsequent improved evaluation of perceived regional leads and lags in climate palaeoceanographic processes will lead to an improved understanding of rapid climate change.

We also demonstrate that dual  $^{14}\text{C}$  and stable isotope ( $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$ ) measurement on single foraminifera can produce temporally accurate benthic ocean chemistry data that is independent of the age-depth paradigm. This development opens up many new avenues in late-glacial palaeoceanographic research, specifically for the vast low SAR areas of the ocean (<10 cm/ka; Fig. 1) that are inaccessible for research using existing methods, thus filling in large spatial gaps in the global, late-glacial climate record. The resulting improvements in spatiotemporal reconstructions of global benthic ventilation conditions of the ocean across the glacial-interglacial transition will help to better understand the interaction between the atmosphere and ocean during periods of rapid climate change.

Retrieving the entire deglacial signal from low SAR sites using our proposed dual  $^{14}\text{C}$  and stable isotope method may also prove to be cost effective, seeing as less elaborate sediment retrieval methods are necessary. Only the top 10-20 cm of the sediment archive are required and the preservation of sediment superposition is not of importance, so it may be possible to retrieve suitable sediment from the low SAR areas of the ocean simply by using grab samples, including those already present in institutional archives. Furthermore, the method we used to  $^{14}\text{C}$ -analyse single foraminifera is efficient and cost-effective, for two main reasons: (1) the elimination of the graphitisation process reduces labour and material costs; (2) the very small sample mass means that the required AMS machine analysis time is greatly reduced.

**Author contributions:** BCL and BM designed the study. BM picked and cut suitable foraminifera tests. LW and BCL carried out  $^{14}\text{C}$  dating. BM, USN and BCL carried out stable isotope analysis. BCL analysed the data and wrote the manuscript with input from the co-authors.

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foraminifera from core T86-10P. Vertical error bars denote  $1\sigma$  error measurement. Horizontal error bars denote the 68.27% highest posterior density (HPD) interval of the calibrated  $^{14}\text{C}$  age (see ~~method~~Method for  $^{14}\text{C}$  calibration process). Also shown are previously published multi-specimen  $\delta^{18}\text{O}$  data from a nearby high SAR (20 cm/ka) record (Repschläger et al., 2015). Vertical error bars represent  $1\sigma$  measurement error. (D) Same as for panel C, but with  $\delta^{13}\text{C}$  instead of  $\delta^{18}\text{O}$ .

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