

Technical note: Large offsets between different datasets of sea water isotopic composition: an illustration of the need to reinforce intercalibration efforts

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19 Abstract 20 We illustrate offsets in seawater isotopic composition between the data sets presented in two 21 recent studies and the LOCEAN seawater isotopic composition dataset, as well as other data 22 for the same years and same regions. These comparisons are carried in the surface waters, for 23 one in the North and South Atlantic, and for the other in the subtropical South-East Indian 24 Ocean. The observed offsets between data sets which exceed 0.10%0 in $\delta^{18}0$ and 0.50%0 in 25 δ²H might in part reflect seasonal or spatial variability. However, they are rather systematic, 26 so they likely originate, at least partially, from different instrumentations and protocols used 27 to measure the water samples. They need to be adjusted in order to ultimately merge the 28 different data sets. This highlights the need to actively share seawater isotopic composition 29 samples dedicated to specific intercomparison of data produced in the different laboratories.





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32	1. Introduction
33	Seawater isotopic composition (180/160 and 2H/1H ratios expressed as δ^{18} 0 and δ^{2} H in
34	‰ in the VSMOW/SLAP scale) is classified as an Essential Ocean/Climate Variable
35	(EOV/ECV) in international programs such as GEOTRACES and GO-SHIP. Stable
36	seawater isotopes (δ^{18} O, δ^{2} H) are used to trace sources of freshwater (precipitation,
37	evaporation, runoff, melting glaciers, sea ice formation and melting), both at the ocean
38	surface and in the ocean interior (Schmidt et al., 2007; Hilaire-Marcel et al., 2021).
39	Except for fractionation during phase changes, the water isotopic composition is nearly
40	conservative in the ocean. A major emphasis is on high latitude oceanography, where
41	continental (or iceberg) glacial melt, formation or melt of sea ice, and high-latitude river
42	inputs (for the Arctic) leave different imprints on the surface ocean isotopic
43	composition. In contrast, few studies have been performed on the isotopic signature in
44	the deep ocean (e.g., Prasanna et al., 2015; Voelker et al., 2015). Seawater isotopes in the
45	upper ocean at low latitudes are often vital for paleoclimatic studies, as they are needed
46	to calibrate proxies of past ocean variability in marine carbonate records such as corals
47	and foraminifera (e.g., PAGES CoralHydro2k working group; Konecky et al., 2020).
48	Seawater isotopes are also important tracers in the coastal ocean, with emphasis on
49	upwelling (Conroy et al., 2014, 2017; Kubota et al., 2022; Lao et al., 2022), and river
50	discharges (e.g., Amazon) (Karr and Showers, 2001). Surface ocean seawater isotopes
51	are also used to characterize evaporation rates and air-sea interactions (Benetti et al.,
52	2017). The isotopic signatures of these different processes are evolving in our warming
53	world, which will imprint on the seawater isotopic composition (Oppo et al., 2007).
54	Additionally, seawater isotope data provide model boundary conditions and allow the
55	assessment of model performance in isotope-enabled Earth system models (e.g. Schmidt
56	et al., 2007; Brady et al., 2019; Cauquoin et al., 2019), thereby improving climate model
57	projections of the future.
58	Stable seawater isotope data have thus been massively produced in the last decades by
59	a variety of methods. For example, most data compiled in the "GISS Global Seawater
60	Oxygen-18 Database -V1.21" for stable seawater isotopes (LeGrande and Schmidt, 2006)

originate from Isotope-ratio Mass Spectrometry (IRMS). They were mostly measured in





62	earlier decades by dual-inlet technology (highest precision), whereas, more recently, the
63	continuous-flow method (lower precision) became widespread for seawater isotope
64	analysis. In the last decade, cavity ring-down spectroscopy (CRDS) turned into another
65	commonly used method as it allows parallel measurement of $\delta^{18}\text{O}$ and $\delta^{2}\text{H}\text{,}$ but with
66	often lower precision (e.g., Voelker et al., 2015). Reverdin et al. (2022) recently compiled
67	a mix of data produced by IRMS and CRDS at LOCEAN
68	(https://www.seanoe.org/data/00600/71186/). As CRDS and other laser techniques
69	(Glaubke et al., 2024) have become more prevalent recently, they contribute a
70	significant part of the new data produced and thus also to the soon to be released
71	CoralHydro2k seawater database for δ^{18} O (δ^{2} H) (focus on the tropics (35°N-35°S);
72	Atwood et al., 2024). There are potential differences between the data produced by the
73	two methods. Typically, CO ₂ -water or H ₂ -water equilibration was used for the IRMS
74	measurements and yields measurements of the activity of water, which decreases with
75	increasing salinity. Furthermore, concentration of divalent cations like Mg** are
76	responsible for slight changes in fractionation factors. On the other hand, the laser
77	methods such as CRDS evaporate the entire sample. If the samples have not been
78	distilled beforehand, there is an issue of salt deposition and of resulting absorption or
79	desorption of water with fractionation effects. In the LOCEAN database (Reverdin et al.,
80	2022), an attempt was made to adjust the data, based on the analysis of Benetti et al
81	(2017b). This was also adopted by at least one other group (Haumann et al., 2022), but
82	overall, there is the possibility of an offset of these data with respect to the ones of other
83	groups using CRDS.
84	It is actually quite common when using water isotope data in studies involving more
85	than one data set, to first evaluate whether there are possible offsets. Intercomparison
86	with earlier data or reference materials was a prerequisite for GEOTRACES sampling
87	campaigns, although for the water isotopes this was, unfortunately, seldomly followed
88	(e.g., Voelker et al., 2015). These intercomparisons often outline systematic differences
89	which could result from the issue outlined above, or from other issues, such as
90	uncertainties in reference materials used, analysis protocols, or isotopic changes in the
91	samples during their handling and storage (Benetti et al., 2017a; Akhoudas et al., 2019;
92	Hennig et al., 2024). In other cases, this was not done, either because the data stood by
93	themselves (Bonne et al., 2019, for δ^{18} 0 and δ^{2} H data), or there was no comparison data





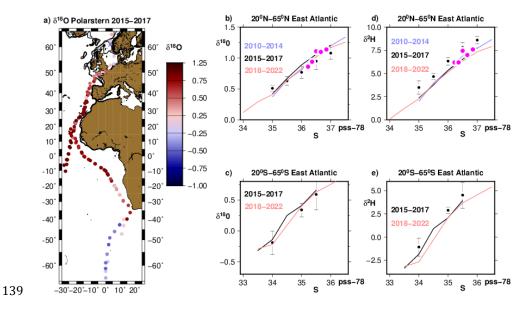
94 available in the same region (Glaubke et al., 2024, for $\delta^{18}O$ data). The possible offsets can 95 however become an issue, when these data are placed in a larger context. For example, Glaubke et al. (2024) identify a large difference in the S- δ^{18} O relationship in the 96 97 subtropical Indian Ocean between their data in the south-eastern part and other data in 98 the south-western Indian Ocean. They also discuss and question differences in the deep 99 water-masses isotopic values between separate data sets, but as these might also be 100 explained by large uncertainties in these data, we will not address them further. 101 Using these two examples (Bonne et al., 2019; Glaubke et al., 2024), the aim of this note 102 is to point out the interest when producing a new data set, of exchanging collected 103 samples to carry a direct comparison, or, if this was not done, to compare the data with 104 other published data and evaluate potential systematic differences. 105 2. Comparisons 106 For identifying possible offsets, we consider surface ocean subsets of the LOCEAN data 107 base in specific regions for roughly the same years as the other data collected. The data 108 extracted are from the same regions as in the datasets of the two studies and are 109 gathered in S- δ^{18} O space as well as in S- δ^{2} H space (available only for the Bonne et al 110 (2019) data set), where S is reported as a practical salinity with the practical salinity 111 scale of 1978 (pss). The assumption done here as in many papers is that the S- δ^{18} O 112 relationship holds on fairly large scales in the surface layer (for the eastern subtropical 113 North Atlantic, see for example, the discussion in Voelker et al (2015) and in Benetti et 114 al. (2017a)). Obviously, this has limitations, such as in areas influenced by more than 115 one water mass or by multiple freshwater end-members (meteoric, continental run-off, 116 sea ice melt or formation, evaporation). 117 2.1 Daily surface data collected from R.V. Polarstern 118 The surface seawater samples originated from daily collection during two years on 119 board RV Polarstern in 2015-2017 (Bonne et al., 2019). There is no salinity provided 120 with the data, and here we chose to associate them with the simultaneously collected 121 thermosalinograph (TSG) data collected on board the RV Polarstern and available from 122 PANGAEA (for each cruise, an indexed file with title starting by 'Continuous 123 thermosalinograh oceanography along Polarstern' is included in PANGAEA: for example,





TSG data for the first cruise (PS90) associated with the isotopic seawater data are found at https://doi.org/10.1594/PANGAEA.858885). The water samples were not collected from the same water line and pumping depth as the TSG data, which can result in differences. This is however likely to be small in most circumstances away from large freshwater input at the sea surface, such as from melting sea ice, intense rainfall and river estuaries. We also applied an adjustment of +0.25% to the δ^{18} O data of Bonne et al. (2019), based on post-analysis identification of a bias in an internal reference material.

We then estimate averages of all the data as a function of salinity in two domains extending poleward of the subtropical salinity maximum toward the higher latitudes in the eastern part of the Atlantic Ocean (thus, 20°N to 65°N and the same in the southern hemisphere). This is done by sorting out the data by salinity classes of 0.5. The LOCEAN data until 2016 in the North and tropical Atlantic were presented in Benetti et al (2017a), showing the tightness of the S- δ^{18} O and S- δ^{2} H relationships in vast domains of the eastern Atlantic. In the North Atlantic, LOCEAN data have been continuously collected since 2011, and south of 10°S in the eastern Atlantic mostly since 2017.







142 Figure 1: Comparison to Bonne et al. (2019). (a) map of RV Polarstern original data set points 143 in eastern Atlantic Ocean east of 30°W. Water isotopes-S scatter diagrams averaged as a 144 function of salinity in 0.5 pss salinity bins (left for δ^{18} 0, and right for δ^{2} H), top for the northern hemisphere and bottom for the southern hemisphere, east of 30°W and outside of 145 146 [20°N, 20°S]. The colored curves represent average relationships of water isotopes in the 147 LOCEAN data base as a function of salinity for three different period ranges, whereas the 148 black dots with error bars are the binned averages of the Bonne et al. (2019) RV Polarstern 149 data in 2015-2017 (after adjustment of +0.25% to δ^{18} O), with the root mean square of the 150 variance reported as error bars. Five individual surface points from Voelker et al (2023) are 151 also plotted (magenta dots). 152 The average relationships found in the LOCEAN data set for three periods overlay well 153 in particular in the northern hemisphere. Uncertainties on individual curves (not 154 shown) are estimated based on the scatter of individual data in each salinity bin. They 155 are typically on the order of 0.01-0.02 (0.05-0.10) % for δ^{18} O (δ^{2} H) respectively in the 156 northern hemisphere (top panel), and a little larger for the less sampled southern 157 hemisphere curves in 2015-2017. Sampling is usually also insufficient at the low end of 158 the salinity range, to reliably estimate an uncertainty. Thus, these different curves nearly 159 overlay within the sampling uncertainty. Five surface samples that were collected in the 160 Northeast Atlantic during the same years within the same salinity range (Voelker et al., 161 2023), also fit well on the North Atlantic curves. The adjusted δ^{18} O data from Bonne et 162 al. (2019) are slightly shifted downward with respect to the curves (Fig. 1b, c), with the 163 plotted standard deviation of individual data around the average not overlapping the 164 LOCEAN data average curves in most cases for the same years 2015-2017. The situation 165 is opposite for the 35 pss bin in the northern hemisphere, with the adjusted δ^{18} O data 166 from Bonne et al. (2019) being above the three LOCEAN average curves, which might be 167 due to samples collected uniquely in the English Channel and North Sea by RV 168 Polarstern in this salinity range, whereas sampling is more geographically-spread in the 169 LOCEAN data base. A difference is also found for $\delta^2 H$, with LOCEAN data been lower than 170 δ^2 H from Bonne et al. (2019) (Fig. 1d, e).

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Glaubke et al. (2024) describe a synthesis of water isotope data in the southern Indian Ocean combining their new dataset in the southeastern Indian Ocean (CROCCA-2S) with earlier data in the south-western Indian Ocean, in particular from LOCEAN, as well as data from the south Australian shelf collected mostly in 2010 (Richardson et al., 2019), and in the equatorial Indian Ocean (Kim et al., 2021). In the most recent version of the LOCEAN data set, in addition to data included in Glaubke et al. (2024) for comparison and collected mostly west of 80°E, there are two transects with surface data through the southeastern Indian Ocean, one collected in February 2017, and the other in March 2024, thus in mid to late austral summer. These transects cross the region covered by the CROCCA-2S data set, albeit not close to west Australia, as well as the area of the Richardson et al. (2019) data set, south of Australia. The LOCEAN data set also contains surface data south of Tasmania (in 2017, as well as in 2020 to 2024). All these data correspond to samples analyzed on a CRDS Picarro L2130 at LOCEAN, and with the protocols discussed in Reverdin et al. (2022). The bottles in which the samples were stored were the same for all the samples, and time between collection and analysis varied, but was mostly on the order of 6 months or less. Thus, this is a rather homogeneously produced set of data in for the years 2016-2024, which spatially overlaps with the data used in Glaubke et al. (2024) collected south of Australia and in the southeastern Indian Ocean (Fig. 2).

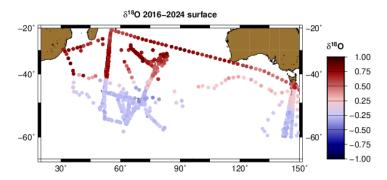


Figure 2: Map of δ^{18} O surface data in the LOCEAN archive for 2016-24, north of 60°S. All these data are associated with S and δ^{2} H data.

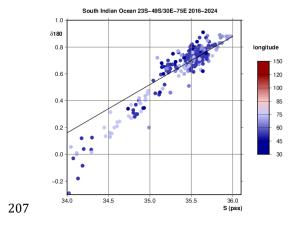
The LOCEAN data distribution indicates some scatter in the S- δ^{18} O distribution in the southwestern Indian Ocean (Fig. 3a) for S larger than 35 pss. Data above the regression





line on Fig. 3a, established for all data with S between 35 and 36 pss, are present only for S larger than 35.0 pss, and are found north of 28°S and in the far south-western Indian Ocean, but with some remnants found all the way to the core of the subtropical gyre near 75°E/35°S (Fig. 3b). Data below the regression line contain most of the data south of 28°S and east of 60°E and connect the salinity maximum region with the lower salinity south of the subtropical front and down to the region south of the polar front (Fig. 3c). These subtropical lower values in S- δ^{18} O space, which appear in the repeated French OISO cruises (in 1998-2024) at 50°E, albeit not all the time, dominate east of 60°E.

206 a)



208 b) c)

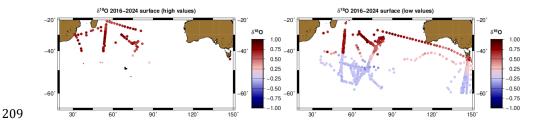


Figure 3: (a) scatter diagram of (S, δ^{18} O) LOCEAN data within the southwestern region (30-75°E/23-49°S) coloured as a function of longitude, with the regression line of the data having a salinity between 35 and 36 pss (black line) overlaid. The spatial distributions of the





213 LOCEAN data with higher and lower δ^{18} O relative to that regression line in the whole Indian

Ocean north of 60°S are shown on panels (b) and (c), respectively.

When focusing on the lower part of the distribution in S- δ^{18} O space (Fig. 3c), one observes a gradual lowering of δ^{18} O from west to east for salinities above 35 (Fig. 4) all the way to 150°E. This lowering is on the order of 0.15 at most, even for the higher salinities (35.5 or more) for which it is strongest (Fig. 4).

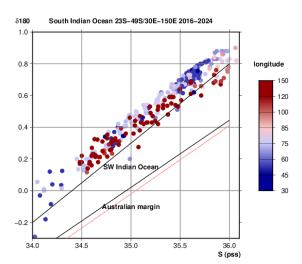


Figure 4: The LOCEAN data below the regression line of Fig. 3a (the ones mapped in Fig. 3c) in S- δ^{18} O space, color-coded as a function of longitude. The two linear relationships recommended in this region between 23°S and 49°S by Glaubke et al. (2024) for the southwest Indian Ocean and for the Australian margin (south of Australia) (we use the original δ^{18} O = 0.4231 * S - 14.7876, instead of the rounded-up relation reported in the paper; R. H. Glaubke, pers. Comm., 2024) are also plotted (black lines), as well as the earlier linear relationship for the 0-600m layer along the Australian margin by Richardson et al. (2018) (in pink).

Thus, besides some gradual and smaller changes, we do not observe in the LOCEAN surface dataset a large sudden change in the (S, δ^{18} O) distribution near 75°E or 85°E between the southeastern and southwestern Indian Ocean, nor a further strong change closer to the Australian coastal margin, as suggested by figures 6 and 7 of Glaubke et al. (2024). Most of the LOCEAN (S, δ^{18} O) data south of 28°S correspond to the mixing of a





233 low salinity end-member characteristic of the fresh waters of the Southern Ocean (at S < 234 34 pss) with waters which are imprinted by air-sea exchange of water in a wider range 235 of values at S > 36 pss, as discussed in Glaubke et al (2024). These LOCEAN (S, δ^{18} O) 236 values are significantly above the linear relationships proposed by Glaubke et al. (2024), 237 in particular for the Australian coastal margins. Furthermore, the LOCEAN data support 238 the presence of a secondary low salinity end member at S < 35 pss with heavier isotopic 239 composition, contributing to the water mass properties in the far southwestern Indian 240 Ocean as well as for the area sampled between 20°S and 28°S north of the subtropical 241 salinity maximum. This could be a contribution of the Indonesian Through Flow and 242 tropical western Indian Ocean surface waters, as discussed by Kim et al. (2021) and 243 Glaubke et al. (2024). 244 3. Discussion 245 In the two cases, we find significant differences between the data sets compared. In the 246 case of the RV Polarstern dataset (Bonne et al., 2019), an error in a specified reference 247 material value was found, and the adjusted data present only small offsets with respect 248 to LOCEAN data, that are slightly negative for δ^{18} O and slightly positive for δ^{2} H. 249 Differences might arise from spatial differences. For example, in the northern 250 hemisphere, values at salinity close to 35 pss mostly originate from the North Sea and 251 English Channel in the Polarstern dataset, thus with more mid-latitude continental 252 influence than for most of the LOCEAN data in the same salinity range which have a 253 contribution of more depleted subpolar and polar freshwater. One expects a larger 254 scatter in the South Atlantic for salinities less than 35 pss, due to intermittent presence 255 of sea ice or iceberg melt, and at higher salinities due to the presence of different water 256 masses originating from the South Atlantic and southeastern Indian Ocean. However, the 257 current data set is not sufficient to estimate it. 258 Furthermore, different seasons were sampled in the two datasets. In the northeastern 259 Atlantic sector, Bonne et al. (2019) surface data east of 30°W were collected in in April 260 and November north of 10°S and in November south of 10°S in the southeastern 261 Atlantic. These data do not suggest large seasonal differences in the Northeast Atlantic, 262 concurring with the LOCEAN (S, δ^{18} O) data in the tropics to mid-latitudes (20 to 50°N), which are tightly distributed along a mean S- δ^{18} O relationship, and thus with low 263





264 seasonal variability (Benetti et al., 2017a; Voelker et al., 2015). The LOCEAN data are 265 not numerous enough in the South-East Atlantic to further evaluate whether the offset is 266 constant throughout the data set, or presents a component related to geophysical 267 temporal or spatial variability. 268 When considering the South Indian Ocean, Glaubke et al. (2024) combined data sets that 269 were processed in different institutes, and potential offsets between those could cause 270 the differences in spatial variability. In particular, Glaubke et al. (2024) outline large 271 spatial contrasts in the S- δ^{18} O relationship across the surface subtropical Indian Ocean 272 and southern Australia that are not observed in the recent version of the LOCEAN 273 database. Seasonal or interannual variability might also contribute to the differences 274 shown on Fig. 3, as the data in the southeastern Indian Ocean from Glaubke et al. (2024 275 were collected in November-December, whereas the data in the LOCEAN database in 276 this region are mostly from February-March. However, at least south of Tasmania, 277 where the LOCEAN data base also contains December data, it does not seem that the 278 seasonal cycle would cause differences larger than 0.05 \(\) at the same salinity. A 279 difference due to seasonality is thus barely identifiable, noting the possible presence of 280 interannual variability and that 0.05 % is the long-term accuracy in the analyses in 281 some centers, such as AWI Potsdam and LOCEAN. Richardson et al. (2018) also 282 commented that there was little difference between a southern winter cruise and March 283 data south of Australia. Further west, near 55-70°E, earlier surface data in the OISO 284 surveys, as well as the vertical upper profiles of OISO station data also suggest a rather 285 modest seasonal variability on the order of 0.10%. Changes could also arise from 286 interannual variability, but the range of interannual variability in the LOCEAN data base 287 is smaller than the difference between the Glaubke et al (2024) curves for the 288 southeastern Indian Ocean and south of Australia and the corresponding LOCEAN data. 289 Thus, a likely contribution for the large differences would be the existence of systematic 290 offsets between the South Indian Ocean/Australia margin data produced in three 291 different institutes that were combined in the Glaubke et al. (2024) study. 292 4. Conclusions 293 What these two comparisons suggest is that offsets are present between different recent 294 data sets published, which exceed 0.10% in $\delta^{18}O$ and 0.50% in $\delta^{2}H$, thus larger than the





295 target long-term accuracy of analyses in individual isotopic laboratories. Moreover, 296 errors in reference material values are always possible and require some post-analysis 297 intercomparisons, such as the one that led to the correction of the RV Polarstern Bonne 298 et al. (2019) data set. Furthermore, one contribution to a systematic difference between 299 the LOCEAN data set and data from other institutes is that the LOCEAN data are 300 reported in concentration scale (Benetti et al., 2017b), thus equivalent to 'freshwater'. 301 The use of the concentration scale corrects possible effects of salt in the water activity 302 measured by IRMS and the effect of salt accumulation during evaporation in laser 303 spectroscopy, which both can lead to fractionation, possibly of similar magnitude 304 (Walker et al., 2016). Different comparisons based on duplicates collected during cruises 305 suggest that this is a main cause of difference between LOCEAN data and other data sets 306 (LOCEAN δ^{18} O data been more positive). Poor conservation of the samples during 307 storage, analytical protocols, or uncertainties in the specified values of reference 308 material are other sources of differences between data produced in different institutes. 309 The methods to carry the intercomparisons and detect systematic differences between 310 different data sets are not numerous. On one hand, one could compare values obtained 311 in specific water masses, for which we expect little variability of the water isotopic 312 composition. This is often used, but such data are not always available, and the resulting 313 uncertainties are difficult to assess. One could also develop a method based on the 314 systematic comparison of nearby data, as is suggested in Fig. 1 when comparing the S-315 water isotopes surface distribution in the North and South Atlantic in the LOCEAN and 316 the RV Polarstern (Bonne et al., 2019) data sets. This could be further improved, but 317 requires that there are enough overlapping data within regions of relatively 318 homogeneous signals. As the data density is not always sufficient, these approaches may 319 fail. Thus, an important alternative approach is to actively share well-preserved water 320 samples, distributed quickly, and dedicated to specific intercomparison of data 321 produced in the different laboratories, building on previous efforts for δ^{13} C-DIC (Cheng 322 et al., 2019). This, together with establishing well-accepted systematic guidelines for 323 data production and quality control, and enhancing scientific exchange between the 324 different institutes needs to be actively pursued, in order to reduce the errors when 325 merging different datasets and increase the potential use of the water isotope data as 326 EOV/ECCVs. Without this effort, the usefulness of the isotopic data for different





327 oceanographic and climate studies is strongly reduced, for example resulting in large 328 uncertainties when establishing different S- δ^{18} O (or S- δ^{2} H) relationships to validate 329 studies of proxies to support paleo-climate reconstructions. 330 331 Data availability 332 The LOCEAN data are available at https://www.seanoe.org/data/00600/71186/. 333 The isotopic data of the Bonne et al. (2019) are available as inidicated in the paper, with 334 here S added, as described in the text from the PANGAEA archive. The Glaubke et al. 335 (2024) data are available as described in the paper. However, among the data used in 336 this paper, we could not access the data from the Richardson et al. (2019) paper. 337 338 Author contribution: GR initiated the study and prepared the manuscript with 339 contributions from all coauthors. AV initiated the intercomparison effort, and AV, CW, 340 and HM contributed to editing the paper. HM was also responsible from producing the 341 data in the Bonne etl. (2019) paper. 342 343 Competing interests: The authors declare that they have no conflict of interest. 344 345 Acknowledgments 346 The LOCEAN isotopic laboratory is supported by OSU Ecce Terra of Sorbonne Université. 347 We are thankful to Catherine Pierre and Jérôme Demange who have set and help run the 348 facility, such as, and for Aïcha Naamar, Marion Benetti and Camille Akhoudas to have 349 measured some of the water samples. We are grateful for support by INSU for samples 350 during the OISO cruises on RV MD2, and by IPEV during the SOCISSE program on RV 351 Astrolabe, with on board support by Patrice Bretel and Rémi Foletto. A. Voelker thanks J. 352 Waniek (IOW, Germany) for collecting the NE Atlantic water samples and R. van Geldern 353 (GeoZentrum Nordbayern, Germany) for analyzing them. She, also, acknowledges 354 financial support by Fundação para a Ciência e a Tecnologia (FCT) through projects 355 Centro de Ciências do Mar do Algarve (CCMAR) basic funding UIDB/04326/2020 356 (https://doi.org/10.54499/UIDB/04326/2020) and programmatic funding UIDP/04326/2020 (https://doi.org/10.54499/UIDP/04326/2020) and the CIMAR 357 358 associated laboratory funding LA/P/0101/2020 359 (https://doi.org/10.54499/LA/P/0101/2020). The RV Polarstern data set was funded





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