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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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Abstract

We illustrate offsets in seawater isotopic composition between the data sets presented in two recent studies and the LOCEAN seawater isotopic composition dataset, as well as other data for the same years and same regions. These comparisons are carried in the surface waters, for one in the North and South Atlantic, and for the other in the subtropical South-East Indian Ocean. The observed offsets between data sets which exceed 0.10‰ in $\delta^{18}O$ and 0.50‰ in $\delta^{2}H$ might in part reflect seasonal or spatial variability. However, they are rather systematic, so they likely originate, at least partially, from different instrumentations and protocols used to measure the water samples. They need to be adjusted in order to ultimately merge the different data sets. This highlights the need to actively share seawater isotopic composition samples dedicated to specific intercomparison of data produced in the different laboratories.

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1. Introduction

Seawater isotopic composition ($^{18}O/^{16}O$ and $^{2}H/^{1}H$ ratios expressed as $\delta^{18}O$ and $\delta^{2}H$ in ‰ in the VSMOW/SLAP scale) is classified as an Essential Ocean/Climate Variable (EOV/ECV) in international programs such as GEOTRACES and GO-SHIP. Stable seawater isotopes ($\delta^{18}O$, $\delta^{2}H$) are used to trace sources of freshwater (precipitation, evaporation, runoff, melting glaciers, sea ice formation and melting), both at the ocean surface and in the ocean interior (Schmidt et al., 2007; Hilaire-Marcel et al., 2021). Except for fractionation during phase changes, the water isotopic composition is nearly conservative in the ocean. A major emphasis is on high latitude oceanography, where continental (or iceberg) glacial melt, formation or melt of sea ice, and high-latitude river inputs (for the Arctic) leave different imprints on the surface ocean isotopic composition. In contrast, few studies have been performed on the isotopic signature in the deep ocean (e.g., Prasanna et al., 2015; Voelker et al., 2015). Seawater isotopes in the upper ocean at low latitudes are often vital for paleoclimatic studies, as they are needed to calibrate proxies of past ocean variability in marine carbonate records such as corals and foraminifera (e.g., PAGES CoralHydro2k working group; Konecky et al., 2020). Seawater isotopes are also important tracers in the coastal ocean, with emphasis on upwelling (Conroy et al., 2014, 2017; Kubota et al., 2022; Lao et al., 2022), and river discharges (e.g., Amazon) (Karr and Showers, 2001). Surface ocean seawater isotopes are also used to characterize evaporation rates and air-sea interactions (Benetti et al., 2017). The isotopic signatures of these different processes are evolving in our warming world, which will imprint on the seawater isotopic composition (Oppo et al., 2007). Additionally, seawater isotope data provide model boundary conditions and allow the assessment of model performance in isotope-enabled Earth system models (e.g. Schmidt et al., 2007; Brady et al., 2019; Cauquoin et al., 2019), thereby improving climate model projections of the future.

Stable seawater isotope data have thus been massively produced in the last decades by a variety of methods. For example, most data compiled in the "GISS Global Seawater 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

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earlier decades by dual-inlet technology (highest precision), whereas, more recently, the continuous-flow method (lower precision) became widespread for seawater isotope analysis. In the last decade, cavity ring-down spectroscopy (CRDS) turned into another commonly used method as it allows parallel measurement of $\delta^{18}O$ and $\delta^{2}H$, but with often lower precision (e.g., Voelker et al., 2015). Reverdin et al. (2022) recently compiled a mix of data produced by IRMS and CRDS at LOCEAN (https://www.seanoe.org/data/00600/71186/). As CRDS and other laser techniques (Glaubke et al., 2024) have become more prevalent recently, they contribute a significant part of the new data produced and thus also to the soon to be released CoralHydro2k seawater database for $\delta^{18}O$ ($\delta^{2}H$) (focus on the tropics (35°N-35°S); Atwood et al., 2024). There are potential differences between the data produced by the two methods. Typically, CO_2-water or H_2-water equilibration was used for the IRMS measurements and yields measurements of the activity of water, which decreases with increasing salinity. Furthermore, concentration of divalent cations like Mg^{++} are responsible for slight changes in fractionation factors. On the other hand, the laser methods such as CRDS evaporate the entire sample. If the samples have not been distilled beforehand, there is an issue of salt deposition and of resulting absorption or desorption of water with fractionation effects. In the LOCEAN database (Reverdin et al., 2022), an attempt was made to adjust the data, based on the analysis of Benetti et al (2017b). This was also adopted by at least one other group (Haumann et al., 2022), but overall, there is the possibility of an offset of these data with respect to the ones of other groups using CRDS.

It is actually quite common when using water isotope data in studies involving more than one data set, to first evaluate whether there are possible offsets. Intercomparison with earlier data or reference materials was a prerequisite for GEOTRACES sampling campaigns, although for the water isotopes this was, unfortunately, seldomly followed (e.g., Voelker et al., 2015). These intercomparisons often outline systematic differences which could result from the issue outlined above, or from other issues, such as uncertainties in reference materials used, analysis protocols, or isotopic changes in the samples during their handling and storage (Benetti et al., 2017a; Akhoudas et al., 2019; Hennig et al., 2024). In other cases, this was not done, either because the data stood by themselves (Bonne et al., 2019, for $\delta^{18}O$ and $\delta^{2}H$ data), or there was no comparison data

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available in the same region (Glaubke et al., 2024, for $\delta^{18}O$ data). The possible offsets can 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-$\delta^{18}O$ relationship in the subtropical Indian Ocean between their data in the south-eastern part and other data in the south-western Indian Ocean. They also discuss and question differences in the deep water-masses isotopic values between separate data sets, but as these might also be explained by large uncertainties in these data, we will not address them further.

Using these two examples (Bonne et al., 2019; Glaubke et al., 2024), the aim of this note is to point out the interest when producing a new data set, of exchanging collected samples to carry a direct comparison, or, if this was not done, to compare the data with other published data and evaluate potential systematic differences.

2. Comparisons

For identifying possible offsets, we consider surface ocean subsets of the LOCEAN data base in specific regions for roughly the same years as the other data collected. The data extracted are from the same regions as in the datasets of the two studies and are gathered in S-$\delta^{18}O$ space as well as in S-$\delta^{2}H$ space (available only for the Bonne et al (2019) data set), where S is reported as a practical salinity with the practical salinity scale of 1978 (pss). The assumption done here as in many papers is that the S-$\delta^{18}O$ relationship holds on fairly large scales in the surface layer (for the eastern subtropical North Atlantic, see for example, the discussion in Voelker et al (2015) and in Benetti et al. (2017a)). Obviously, this has limitations, such as in areas influenced by more than one water mass or by multiple freshwater end-members (meteoric, continental run-off, sea ice melt or formation, evaporation).

2.1 Daily surface data collected from R.V. Polarstern

The surface seawater samples originated from daily collection during two years on board RV Polarstern in 2015-2017 (Bonne et al., 2019). There is no salinity provided with the data, and here we chose to associate them with the simultaneously collected thermosalinograph (TSG) data collected on board the RV Polarstern and available from PANGAEA (for each cruise, an indexed file with title starting by 'Continuous thermosalinograh oceanography along Polarstern' is included in PANGAEA: for example,

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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 $\delta^{18}O$ 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-$\delta^{18}O$ and S-$\delta^{2}H$ 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.

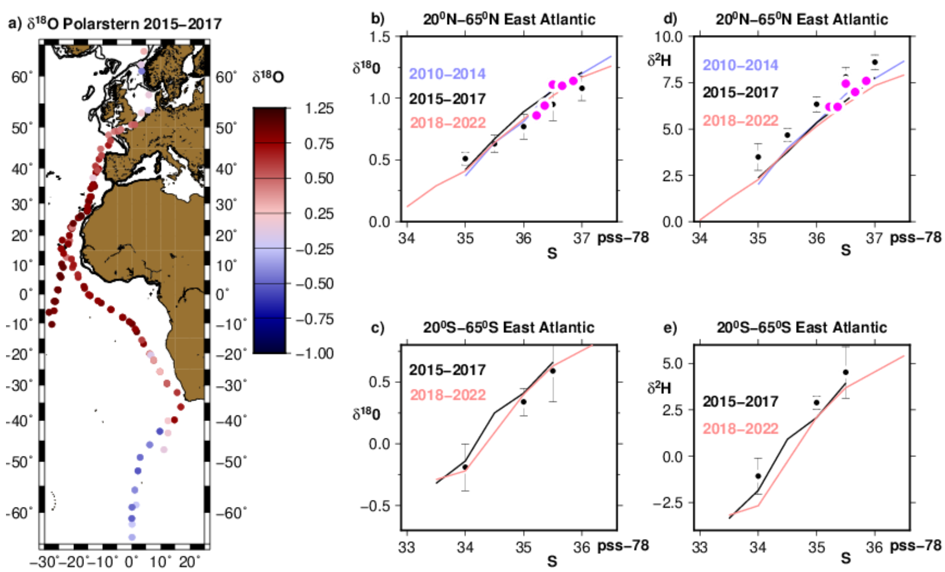

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Figure 1: Comparison to Bonne et al. (2019). (a) map of RV Polarstern original data set points in eastern Atlantic Ocean east of 30°W. Water isotopes-S scatter diagrams averaged as a function of salinity in 0.5 pss salinity bins (left for $\delta^{18}O$, and right for $\delta^{2}H$), top for the northern hemisphere and bottom for the southern hemisphere, east of 30°W and outside of [20°N, 20°S]. The colored curves represent average relationships of water isotopes in the LOCEAN data base as a function of salinity for three different period ranges, whereas the black dots with error bars are the binned averages of the Bonne et al. (2019) RV Polarstern data in 2015-2017 (after adjustment of +0.25‰ to $\delta^{18}O$), with the root mean square of the variance reported as error bars**.** Five individual surface points from Voelker et al (2023) are also plotted (magenta dots).

The average relationships found in the LOCEAN data set for three periods overlay well in particular in the northern hemisphere. Uncertainties on individual curves (not shown) are estimated based on the scatter of individual data in each salinity bin. They are typically on the order of 0.01-0.02 (0.05-0.10) ‰ for $\delta^{18}O$ ($\delta^{2}H$) respectively in the northern hemisphere (top panel), and a little larger for the less sampled southern hemisphere curves in 2015-2017. Sampling is usually also insufficient at the low end of the salinity range, to reliably estimate an uncertainty. Thus, these different curves nearly overlay within the sampling uncertainty. Five surface samples that were collected in the Northeast Atlantic during the same years within the same salinity range (Voelker et al., 2023), also fit well on the North Atlantic curves. The adjusted $\delta^{18}O$ data from Bonne et al. (2019) are slightly shifted downward with respect to the curves (Fig. 1b, c), with the plotted standard deviation of individual data around the average not overlapping the LOCEAN data average curves in most cases for the same years 2015-2017. The situation is opposite for the 35 pss bin in the northern hemisphere, with the adjusted $\delta^{18}O$ data from Bonne et al. (2019) being above the three LOCEAN average curves, which might be due to samples collected uniquely in the English Channel and North Sea by RV Polarstern in this salinity range, whereas sampling is more geographically-spread in the LOCEAN data base. A difference is also found for $\delta^{2}H$, with LOCEAN data been lower than $\delta^{2}H$ from Bonne et al. (2019) (Fig. 1d, e).

2.2 Southern subtropical Indian Ocean

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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).

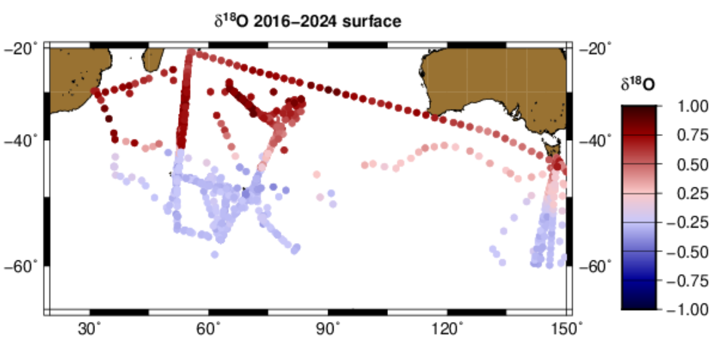

Figure 2: Map of $\delta^{18}O$ surface data in the LOCEAN archive for 2016-24, north of 60°S. All these data are associated with S and $\delta^{2}H$ data.

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

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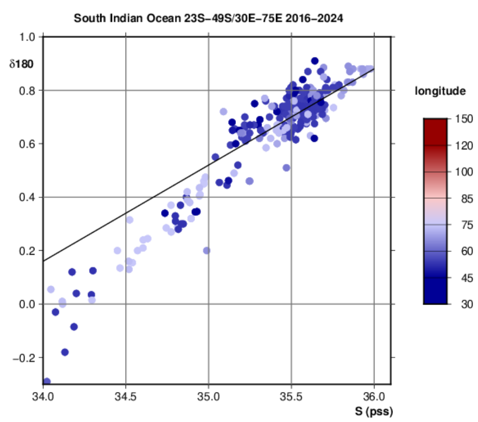
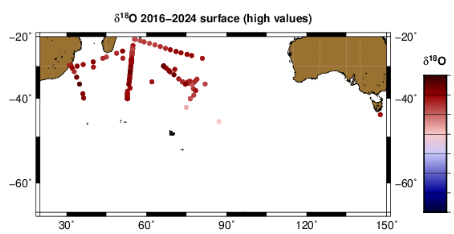
line on Fig. 3a, established for all data with S between 35 and 36 pss, are present only for S larger than 35.0 pss, and are found north of 28°S and in the far south-western Indian Ocean, but with some remnants found all the way to the core of the subtropical gyre near 75°E/35°S (Fig. 3b). Data below 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.

a)



b)



c)

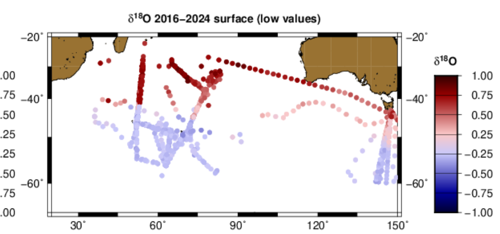


Figure 3: (a) scatter diagram of (S, δ^{18}O) LOCEAN data within the southwestern region (30-75°E/23-49°S) coloured as a function of longitude, with the regression line of the data having a salinity between 35 and 36 pss (black line) overlaid. The spatial distributions of the

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LOCEAN data with higher and lower $\delta^{18}O$ 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-$\delta^{18}O$ space (Fig. 3c), one observes a gradual lowering of $\delta^{18}O$ 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).

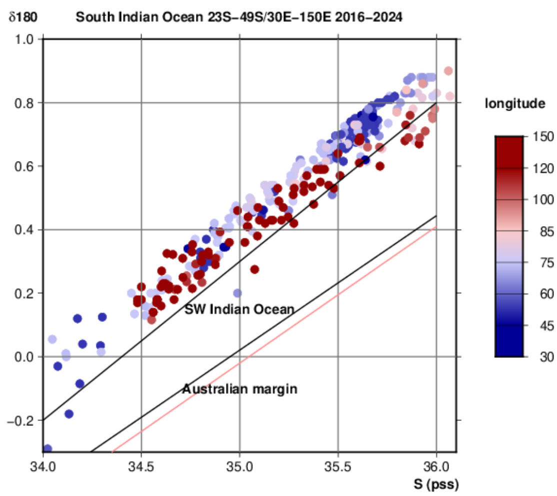

Figure 4: The LOCEAN data below the regression line of Fig. 3a (the ones mapped in Fig. 3c) in S-$\delta^{18}O$ space, color-coded as a function of longitude. The two linear relationships recommended in this region between 23°S and 49°S by Glaubke et al. (2024) for the south-west Indian Ocean and for the Australian margin (south of Australia) (we use the original $\delta^{18}O = 0.4231 * S - 14.7876$, 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, $\delta^{18}O$) 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, $\delta^{18}O$) data south of 28°S correspond to the mixing of a

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low salinity end-member characteristic of the fresh waters of the Southern Ocean (at S < 34 pss) with waters which are imprinted by air-sea exchange of water in a wider range of values at S > 36 pss, as discussed in Glaubke et al (2024). These LOCEAN (S, $\delta^{18}O$) values are significantly above the linear relationships proposed by Glaubke et al. (2024), in particular for the Australian coastal margins. Furthermore, the LOCEAN data support the presence of a secondary low salinity end member at S < 35 pss with heavier isotopic composition, contributing to the water mass properties in the far southwestern Indian Ocean as well as for the area sampled between 20°S and 28°S north of the subtropical salinity maximum. This could be a contribution of the Indonesian Through Flow and tropical western Indian Ocean surface waters, as discussed by Kim et al. (2021) and Glaubke et al. (2024).

3. Discussion

In the two cases, we find significant differences between the data sets compared. In the case of the RV Polarstern dataset (Bonne et al., 2019), an error in a specified reference material value was found, and the adjusted data present only small offsets with respect to LOCEAN data, that are slightly negative for $\delta^{18}O$ and slightly positive for $\delta^{2}H$. Differences might arise from spatial differences. For example, in the northern hemisphere, values at salinity close to 35 pss mostly originate from the North Sea and English Channel in the Polarstern dataset, thus with more mid-latitude continental influence than for most of the LOCEAN data in the same salinity range which have a contribution of more depleted subpolar and polar freshwater. One expects a larger scatter in the South Atlantic for salinities less than 35 pss, due to intermittent presence of sea ice or iceberg melt, and at higher salinities due to the presence of different water masses originating from the South Atlantic and southeastern Indian Ocean. However, the current data set is not sufficient to estimate it.

Furthermore, different seasons were sampled in the two datasets. In the northeastern Atlantic sector, Bonne et al. (2019) surface data east of 30°W were collected in in April and November north of 10°S and in November south of 10°S in the southeastern Atlantic. These data do not suggest large seasonal differences in the Northeast Atlantic, concurring with the LOCEAN (S, $\delta^{18}O$) data in the tropics to mid-latitudes (20 to 50°N), which are tightly distributed along a mean S-$\delta^{18}O$ relationship, and thus with low

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seasonal variability (Benetti et al., 2017a; Voelker et al., 2015). The LOCEAN data are not numerous enough in the South-East Atlantic to further evaluate whether the offset is constant throughout the data set, or presents a component related to geophysical temporal or spatial variability.

When considering the South Indian Ocean, Glaubke et al. (2024) combined data sets that were processed in different institutes, and potential offsets between those could cause the differences in spatial variability. In particular, Glaubke et al. (2024) outline large spatial contrasts in the S-δ^{18}O relationship across the surface subtropical Indian Ocean and southern Australia that are not observed in the recent version of the LOCEAN database. Seasonal or interannual variability might also contribute to the differences shown on Fig. 3, as the data in the southeastern Indian Ocean from Glaubke et al. (2024 were collected in November-December, whereas the data in the LOCEAN database in this region are mostly from February-March. However, at least south of Tasmania, where the LOCEAN data base also contains December data, it does not seem that the seasonal cycle would cause differences larger than 0.05 ‰ at the same salinity. A difference due to seasonality is thus barely identifiable, noting the possible presence of interannual variability and that 0.05 ‰ is the long-term accuracy in the analyses in some centers, such as AWI Potsdam and LOCEAN. Richardson et al. (2018) also commented that there was little difference between a southern winter cruise and March data south of Australia. Further west, near 55-70°E, earlier surface data in the OISO surveys, as well as the vertical upper profiles of OISO station data also suggest a rather modest seasonal variability on the order of 0.10‰. Changes could also arise from interannual variability, but the range of interannual variability in the LOCEAN data base is smaller than the difference between the Glaubke et al (2024) curves for the southeastern Indian Ocean and south of Australia and the corresponding LOCEAN data. Thus, a likely contribution for the large differences would be the existence of systematic offsets between the South Indian Ocean/Australia margin data produced in three different institutes that were combined in the Glaubke et al. (2024) study.

4. Conclusions

What these two comparisons suggest is that offsets are present between different recent data sets published, which exceed 0.10‰ in δ^{18}O and 0.50‰ in δ^{2}H, thus larger than the

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target long-term accuracy of analyses in individual isotopic laboratories. Moreover, errors in reference material values are always possible and require some post-analysis intercomparisons, such as the one that led to the correction of the RV Polarstern Bonne et al. (2019) data set. Furthermore, one contribution to a systematic difference between the LOCEAN data set and data from other institutes is that the LOCEAN data are reported in concentration scale (Benetti et al., 2017b), thus equivalent to 'freshwater'. The use of the concentration scale corrects possible effects of salt in the water activity measured by IRMS and the effect of salt accumulation during evaporation in laser spectroscopy, which both can lead to fractionation, possibly of similar magnitude (Walker et al., 2016). Different comparisons based on duplicates collected during cruises suggest that this is a main cause of difference between LOCEAN data and other data sets (LOCEAN $\delta^{18}O$ data been more positive). Poor conservation of the samples during storage, analytical protocols, or uncertainties in the specified values of reference material are other sources of differences between data produced in different institutes.

The methods to carry the intercomparisons and detect systematic differences between different data sets are not numerous. On one hand, one could compare values obtained in specific water masses, for which we expect little variability of the water isotopic composition. This is often used, but such data are not always available, and the resulting uncertainties are difficult to assess. One could also develop a method based on the systematic comparison of nearby data, as is suggested in Fig. 1 when comparing the S-water isotopes surface distribution in the North and South Atlantic in the LOCEAN and the RV Polarstern (Bonne et al., 2019) data sets. This could be further improved, but requires that there are enough overlapping data within regions of relatively homogeneous signals. As the data density is not always sufficient, these approaches may fail. Thus, an important alternative approach is to actively share well-preserved water samples, distributed quickly, and dedicated to specific intercomparison of data produced in the different laboratories, building on previous efforts for $\delta^{13}C$-DIC (Cheng et al., 2019). This, together with establishing well-accepted systematic guidelines for data production and quality control, and enhancing scientific exchange between the different institutes needs to be actively pursued, in order to reduce the errors when merging different datasets and increase the potential use of the water isotope data as EOV/ECCVs. Without this effort, the usefulness of the isotopic data for different

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oceanographic and climate studies is strongly reduced, for example resulting in large uncertainties when establishing different S-$\delta^{18}O$ (or S-$\delta^{2}H$) relationships to validate studies of proxies to support paleo-climate reconstructions.

Data availability

The LOCEAN data are available at https://www.seanoe.org/data/00600/71186/. The isotopic data of the Bonne et al. (2019) are available as inidicated in the paper, with here S added, as described in the text from the PANGAEA archive. The Glaubke et al. (2024) data are available as described in the paper. However, among the data used in this paper, we could not access the data from the Richardson et al. (2019) paper.

Author contribution: GR initiated the study and prepared the manuscript with contributions from all coauthors. AV initiated the intercomparison effort, and AV, CW, and HM contributed to editing the paper. HM was also responsible from producing the data in the Bonne etl. (2019) paper.

Competing interests: The authors declare that they have no conflict of interest.

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