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# A synthesis of ocean total alkalinity and dissolved inorganic carbon measurements from 1993 to 2022: the SNAPO-CO2-v1 dataset

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**Abstract.** Total alkalinity ( $A_T$ ) and dissolved inorganic carbon ( $C_T$ ) in the oceans are important properties with respect to understanding the ocean carbon cycle and its link to global change (ocean carbon sinks and sources, ocean acidification) and ultimately finding carbon-based solutions or mitigation procedures (marine carbon removal). We present a database of more than 44 400  $A_T$  and  $C_T$  observations along with basic ancillary data (spatiotemporal location, depth, temperature and salinity) from various ocean regions obtained, mainly in the framework of French projects, since 1993. This includes both surface and water column data acquired in the open ocean, coastal zones and in the Mediterranean Sea and either from time series or dedicated one-off cruises. Most  $A_T$  and  $C_T$  data in this synthesis were measured from discrete samples using the same closed-cell potentiometric titration calibrated with Certified Reference Material, with an overall accuracy of  $\pm 4 \mu\text{mol kg}^{-1}$  for both  $A_T$  and  $C_T$ . The data are provided in two separate datasets – for the Global Ocean and the Mediterranean Sea (<https://doi.org/10.17882/95414>, Metzl et al., 2023), respectively – that offer a direct use for regional or global purposes, e.g.,  $A_T$ –salinity relationships, long-term  $C_T$  estimates, and constraint and validation of diagnostic  $C_T$  and  $A_T$  reconstructed fields or ocean carbon and coupled climate–carbon models simulations as well as data derived from Biogeochemical-Argo (BGC-Argo) floats. When associated with other properties, these data can also be used to calculate pH, the fugacity of CO<sub>2</sub> ( $f\text{CO}_2$ ) and other carbon system properties to derive ocean acidification rates or air–sea CO<sub>2</sub> fluxes.

## 1 Introduction

Since 1750, human activities have added 700 ( $\pm 75$ ) PgC of anthropogenic carbon dioxide to the atmosphere by burning fossil fuels, producing cement and changing land use (Friedlingstein et al., 2022), thereby driving up the atmospheric carbon dioxide (CO<sub>2</sub>) level and leading to unequivocal global change. The ocean plays a major role in reducing the impact of climate change by absorbing more than 90 % of the excess heat in the climate system (Cheng et al., 2020; von Schuckmann et al., 2020, 2023; IPCC, 2022) and about 25 % of anthropogenic CO<sub>2</sub> (Friedlingstein et al., 2022). However, oceanic CO<sub>2</sub> uptake changes the chemistry of seawater, reducing its buffering capacity (Revelle and Suess, 1957; Jiang et al., 2023a) and leading to a process known as “ocean acidification”, with potential impacts on marine organisms (Fabry et al., 2008; Doney et al., 2009, 2020; Gattuso et al., 2015). Along with atmospheric CO<sub>2</sub> concentrations, surface-ocean temperature, ocean heat content, sea level, sea ice and glaciers, ocean acidification (a decrease in pH) is now recognized by the World Meteorological Organization (WMO) as one of the seven key global climate indicators (WMO, 2018). In the framework of the 2030 Agenda for Sustainable Development, the United Nations established a set of Sustainable Development Goals (SDGs; United Nations, 2020), including a goal dedicated to the ocean (SDG 14, “Life below water”) which calls for work to “conserve and sustainably use the oceans, seas and marine resources for sustainable development”. Ocean acidification is specifically referred in the SDG indicator 14.3.1 coordinated by the Intergovernmental Oceanographic Commission (IOC) of the United Nations Educational, Scientific and Cultural Organization (UNESCO). Observing the carbonate system in the oceans and marginal seas and understanding how this system changes over time is, thus, highly relevant not only to quantify the

Global Ocean carbon budget, the anthropogenic CO<sub>2</sub> inventories or ocean acidification rates but also to understand and simulate the processes that govern the complex CO<sub>2</sub> cycle in the ocean and to better predict the future evolution of climate and global changes (Eyring et al., 2016; Kwiatkowski et al., 2020; Jiang et al., 2023a).

The number and quality of ocean CO<sub>2</sub> fugacity ( $f\text{CO}_2$ ), total alkalinity ( $A_T$ ), dissolved inorganic carbon ( $C_T$ ) and pH measurements have increased substantially over the past few decades. Quality-controlled observations are now regularly assembled in global data syntheses such as SOCAT (Surface Ocean CO<sub>2</sub> Atlas; Pfeil et al., 2013; Bakker et al., 2014, 2016) and GLODAP (Global Ocean Data Analysis Project; Key et al., 2004; Olsen et al., 2016, 2019, 2020; Lauvset et al., 2021, 2022). These datasets allow for the evaluation of properties’ trends in the Global Ocean, including the change in the ocean CO<sub>2</sub> sink (e.g., Wanninkhof et al., 2013; Friedlingstein et al., 2022; Watson et al., 2020), anthropogenic CO<sub>2</sub> inventories (e.g., Sabine et al., 2004; Khatiwala et al., 2013; Gruber et al., 2019) and ocean acidification (Lauvset et al., 2015, 2020; Jiang et al., 2019; Feely et al., 2023; Ma et al., 2023). Thanks to publicly available, consistent and quality-controlled databases, new methods have been recently developed (Carter et al., 2016; Sauzède et al., 2017; Bittig et al., 2018) to reproduce  $A_T$  and  $C_T$  distributions from other properties, like temperature, salinity and oxygen, that are more often observed in the water column, especially by autonomous floats (Claustre et al., 2020; Mignot et al., 2023). These methods (named CANYON-B and CONTENT; Bittig et al., 2018) are now also used to help inform decisions regarding GLODAP data quality control or to fill in observational gaps (Olsen et al., 2019, 2020; Tanhua et al., 2019, 2021). The GLODAP data products have also been successfully used to construct new Global Ocean  $A_T$  and  $C_T$  climatological monthly fields at the surface and in the water

column using neural network methods (e.g., Broullón et al., 2019, 2020).

Following pioneer works that produced various global-ocean climatologies of the sea-surface carbonate system (Millero et al., 1998; Lee et al., 2000, 2006; Takahashi et al., 2002, 2009, 2014; Sasse et al., 2013; Jiang et al., 2019), the coupling of  $f\text{CO}_2$  data (from SOCAT) and  $A_T$  data (from GLODAP) now enables reconstruction of the full carbonate system in the surface ocean at a monthly timescale to investigate temporal trends at a decadal timescale (e.g., Gregor and Gruber, 2021; Keppler et al., 2023).

International projects such as SOCAT and GLODAP offer an important way to synthesize ocean carbon data. In these projects, each observation is quality controlled, offering users high-quality observations for regional or global analysis, either for process analysis or to constrain or validate ocean and coupled climate–carbon models (Coupled Model Intercomparison Project Phase 6 – CMIP6, e.g., Lerner et al., 2021). SOCAT is a publicly available synthesis product initiated in 2007 (Metzl et al., 2007) for quality-controlled surface-ocean  $f\text{CO}_2$  observations made by the international marine carbon research community (Bakker et al., 2016). The first SOCAT version was released in 2011 (Pfeil et al., 2013; Sabine et al., 2013), followed by six SOCAT versions (Bakker et al., 2014, 2016). The last version in 2023 included more than 40 million  $f\text{CO}_2$  data with an accuracy better than 5  $\mu\text{atm}$  (Bakker et al., 2023). One important component of SOCAT is the use of data to estimate global air–sea  $\text{CO}_2$  fluxes based on reconstructed  $p\text{CO}_2$  fields (e.g., Surface Ocean  $p\text{CO}_2$  Mapping Intercomparison – SOCOM; Rödenbeck et al., 2015). Since 2015, these results have been included in the estimate of the global carbon budget each year (Le Quéré et al., 2015; Friedlingstein et al., 2022).

On the other hand, following the World Ocean Circulation Experiment (WOCE) and Joint Global Ocean Flux Study (JGOFS) era in the 1990s, when almost all observations were started to be synthesized in a specific recommended format (Joyce and Corry, 1994), GLODAP focused on water-column carbon observations (and other properties). Following the original GLODAP data product (Key et al., 2004), the project accumulated many new quality-controlled observations. One important achievement of GLODAP has been the use of data to estimate the anthropogenic  $\text{CO}_2$  inventory or its change over decades (Sabine et al., 2004; Gruber et al., 2019). Both products, SOCAT and GLODAP, are relevant tools to detect oceanic acidification rates (Lauvset et al., 2015; Jiang et al., 2019; Feely et al., 2023; Ma et al., 2023).

Although these projects include many international ocean observations, there are ocean- $\text{CO}_2$ -related observations all around the world (published or unpublished), such as total alkalinity and dissolved inorganic carbon, that are not included in SOCAT nor GLODAP. This is because SOCAT accepts and controls only  $f\text{CO}_2$  data, whereas GLODAP includes and controls water-column data, mainly from WOCE, Global Ocean Ship-based Hydrographic Investigations Pro-

gram (GO-SHIP), and Climate Variability and Predictability Experiment (CLIVAR) cruises. It should be noted that many ocean carbon observations in various formats can be also found in dedicated database, such as the National Centers for Environmental Information (NCEI) Ocean Carbon and Acidification Data System (OCADS) (formerly CDIAC-Ocean; Jiang et al., 2023b, <https://www.ncei.noaa.gov/products/ocean-carbon-acidification-data-system>, last access: 22 December 2023), PANGAEA (<https://www.pangaea.de/>, last access: 22 December 2023) or SEANOE (<https://www.seanoe.org/>, last access: 22 December 2023). In this context, progress in the data synthesis of ocean carbon observations that would offer new high-quality products for the community (e.g., for GOA-ON, <http://www.goa-on.org>, last access: 22 December 2023, IOC/SDG 14.1.3, <https://oa.iode.org/>, last access: 22 December 2023, Tilbrook et al., 2019) is recommended.

In this work, we present a synthesis of more than 44 400  $A_T$  and  $C_T$  observations obtained over the 1993–2022 period during various cruises or at time series' stations mainly supported by French projects. This dataset merges observations measured with the same instruments and is, thus, analytically coherent. Most of the data have an accuracy better than  $\pm 4 \mu\text{mol kg}^{-1}$ , i.e., between the climate ( $\pm 2 \mu\text{mol kg}^{-1}$ ) and weather ( $\pm 10 \mu\text{mol kg}^{-1}$ ) goals (Newton et al., 2015; Bockmon and Dickson, 2015). Hereafter, this dataset will be referred to as SNAPO-CO<sub>2</sub>-v1. We describe the data assemblage and associated quality control and discuss some potential uses of this dataset.

## 2 Data collection

The time series' projects and research cruises during which data were collated are listed in Table 1, with the corresponding references given in the Supplement (Table S1) and the sampling locations displayed in Fig. 1. Sampling was performed either from CTD-rosette casts (Niskin bottles) or from the ship's seawater supply (intake at about 5 m depth, depending on the ship and swell). Samples collected in 500 mL borosilicate glass bottles were poisoned with 100–300  $\mu\text{L}$  of  $\text{HgCl}_2$  (depending on the cruise) and closed with greased stoppers (Apiezon<sup>®</sup>) that were held tight using an elastic band, following the standard operating procedure (SOP) protocol (Dickson et al., 2007). Some samples were also collected in 500 mL bottles closed with screw caps. After the completion of each cruise, discrete samples were returned back to the LOCEAN (Laboratoire d'Océanographie et du Climat: Expérimentations et Approches Numériques) laboratory (Paris, France) and stored in a dark room at 4 °C before analysis; analysis generally took place within 2–3 months of sampling (sometimes within a week). Some samples were also measured for specific process studies on benthic corals (e.g., Maier et al., 2012; McCulloch et al., 2012) or for mesocosm and culture experiments, but these data are

not included in this synthesis because they do not represent the natural ocean state (e.g., the addition of Sahara dust during the “DUst experiment in a low-Nutrient, low-chlorophyll Ecosystem” project – DUNE; Ridame et al., 2014).

As opposed to  $p\text{CO}_2$ , surface  $A_T$  or  $C_T$  observations are generally obtained from discrete sampling (measured aboard a vessel or onshore). Few cruises offer semicontinuous sea-surface  $A_T$  or  $C_T$  observations (e.g., Metzl et al., 2006), but new instrumental developments (Seilmann et al., 2020) have now enabled  $A_T$  measurements on Ship of Opportunity Program (SOOP) lines. In addition to discrete samples analyzed for various projects conducted mainly in the North Atlantic, tropical Atlantic, tropical Pacific, Mediterranean Sea and coastal regions (Table 1), we complemented this synthesis with  $A_T$  and  $C_T$  surface observations obtained in the Indian and Southern oceans during the OISO (Océan Indien Service d’Observation) cruises in 1998–2018 (Metzl et al., 2006; Leseurre et al., 2022; data also available from NCEI OCADS: [https://www.nodc.noaa.gov/ocads/oceans/VOS\\_Program/](https://www.nodc.noaa.gov/ocads/oceans/VOS_Program/), last access: 22 December 2023) and the recent CLIM-EPARSEs cruise conducted in the Mozambique Channel in April 2019 (Lo Monaco et al., 2020, 2021). For OISO cruises, the water-column observations are part of the CARINA (CARbon IN the Atlantic) and GLODAP synthesis products (Lo Monaco et al., 2010; Olsen et al., 2016, 2019, 2020) and are not included here. Except when otherwise specified, all data in this synthesis were obtained using the same technique used in either the laboratory or at sea (for the OISO 1998–2018 and CLIM-EPARSEs 2019 cruises).

### 3 Method, accuracy, repeatability, intercomparison and quality control

#### 3.1 Method and accuracy

Since 2003, the discrete samples returned to the SNAPO-CO<sub>2</sub> service facilities (LOCEAN, Paris) have been simultaneously analyzed for  $A_T$  and  $C_T$  using potentiometric titration with a closed cell (Edmond, 1970; Goyet et al., 1991). The same technique was used at sea for underway surface water measurements during OISO and CLIM-EPARSEs cruises (indicated using the footnote in Table 1). For two time series, the dataset also includes measurements obtained before 2000 using other techniques: the DYFAMED time series’ observations measured between 1998 and 2000 in the Mediterranean Sea (Copin-Montégut and Bégovic, 2002; Coppola et al., 2020) and the SURATLANT time series’ values acquired from 1993 to 1997 in the North Atlantic subpolar gyre (Reverdin et al., 2018). We also include  $A_T$  data from the river La Penzé (Brittany) from 2019 to 2020 (Yann Bozec, Station Biologique de Roscoff – SBR, personal communication, 2020).

In the late 1980s, the so-called “JGOFS-IOC Advisory Panel on Ocean CO<sub>2</sub>” highlighted the need for standard anal-

ysis protocols and for the development of Certified Reference Materials (CRMs) for inorganic carbon measurements (Poisson et al., 1990; UNESCO, 1990, 1991). The CRMs were provided to international laboratories by Andrew Dickson (Scripps Institution of Oceanography, San Diego, USA), starting in 1990 for  $C_T$  and in 1996 for  $A_T$ . Thus, these CRMs were available during this work and were used to calibrate the measurements (CRM batch numbers used for each cruise are listed in Table S2 in the Supplement). The concentrations of the CRMs that we used varied between 2193 and 2426  $\mu\text{mol kg}^{-1}$  for  $A_T$  and between 1968 and 2115  $\mu\text{mol kg}^{-1}$  for  $C_T$ , corresponding to the range of concentrations observed in open-ocean water. The CRM accuracy, as indicated on the certificate for each batch, is around  $\pm 0.5 \mu\text{mol kg}^{-1}$  for both  $A_T$  and  $C_T$  ([https://www.nodc.noaa.gov/ocads/oceans/Dickson\\_CRM/batches.html](https://www.nodc.noaa.gov/ocads/oceans/Dickson_CRM/batches.html), last access: 22 December 2023).

The results of analyses performed on 965 CRM bottles (different batches) between 2013 and 2023 are presented in Fig. 2. The standard deviations of the differences in measurements were around  $\pm 3.5 \mu\text{mol kg}^{-1}$  on average for both  $A_T$  and  $C_T$ . For unknown reasons, the differences were occasionally up to 10–15  $\mu\text{mol kg}^{-1}$  (0.8 % of the data; Fig. S2). These few CRM measurements were discarded with respect to the data processing. On average, excluding some outliers, standard deviations of the differences for 1090 CRM analyses were  $\pm 2.71 \mu\text{mol kg}^{-1}$  for  $A_T$  and  $\pm 2.86 \mu\text{mol kg}^{-1}$  for  $C_T$ . We did not detect any specific signal for CRM analyses (e.g., larger uncertainty depending on the batch number or temporal drifts during analyses; Fig. 2); however, for some cruises, the accuracy based on CRMs could be slightly better than 3  $\mu\text{mol kg}^{-1}$  (e.g., Marrec et al., 2014; Touratier et al., 2016; Ganachaud et al., 2017; Wimart-Rousseau et al., 2020).

#### 3.2 Repeatability

For some projects, duplicates have regularly been sampled (e.g., SOMLIT-Point-B, SOMLIT-BREST and BOUSSOLE/DYFAMED) or replicate bottles have been sampled at selected depths at fixed stations during the cruises (e.g., OUTPACE 2015 and SOMBA 2014). The results of  $A_T$  and  $C_T$  repeatability are synthesized in Table 2. Figure 3 shows an example of regular duplicates from the following time series: SOMLIT-Point-B in the coastal Mediterranean Sea, SOMLIT-BREST in the Bay of Brest in the coastal Iroise Sea and BOUSSOLE/DYFAMED in the Ligurian Sea. For the 26 OISO cruises conducted between 1998 and 2018 and the CLIM-EPARSEs cruise in April 2019, the repeatability was evaluated from duplicate analyses (within 20 min) of continuous underway sea-surface samples at the same location (when the ship was stopped). Similarly to what was found for the CRM measurements (Fig. S2), differences in duplicates are occasionally higher than 10–15  $\mu\text{mol kg}^{-1}$  (Fig. 3), although most of the duplicates for all projects are

**Table 1.** List of cruises in the SNAPO-CO2-v1 dataset. This is organized by region for the Global Ocean and coastal zones and for the Mediterranean Sea (MedSea). The reader is referred to Tables S1, S2, S4 and S4 in the Supplement for a list of the laboratories, the CRMs used, the corresponding references and the DOIs of cruises. Nb denotes the number of data for each cruise or time series.

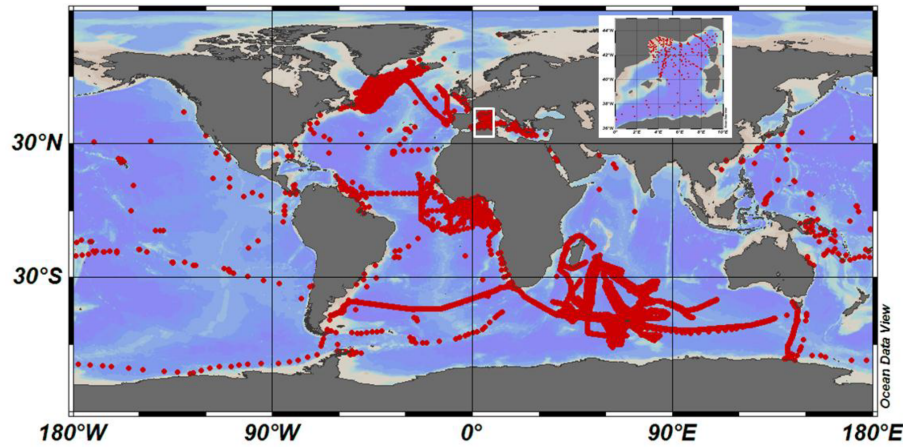
Cruise/Project	Start	End	Region	Sampling	Nb
AWIPEV	2015	2021	Arctic	Surface and subsurface	195
SURATLANT and RREX	1993	2017	North Atlantic	Surface	2832
OVIDE	2006	2018	North Atlantic	Surface and water column	397
STRASSE	2012		North Atlantic	Water column	205
EUREC4A-OA	2020		North Atlantic	Surface and water column	135
PROTEUS	2010		North Atlantic	Water column	27
CHANNEL	2012	2015	English Channel	Surface	696
SOMLIT-BREST	2008	2019	Coastal North Atlantic	Surface	1174
SOMLIT-Roscoff	2009	2019	Coastal North Atlantic	Surface and 60 m	801
ECOSCOPA	2017	2019	Coastal North Atlantic	Surface	67
Penzé	2011	2020	Brittany, river	Surface and subsurface	148
Aulne	2009	2010	Brittany, river	Surface	27
Élorn	2009	2009	Brittany, river	Surface	28
BIOZAIRE	2003	2004	Tropical Atlantic	Water column	87
EGEE	2005	2007	Tropical Atlantic	Surface	199
PIRATA-FR	2009	2017	Tropical Atlantic	Surface and water column	513
PLUMAND	2007		Tropical Atlantic	Surface	38
OUTPACE	2015		Tropical Pacific	Water column	240
PANDORA	2012		Solomon Sea	Water column	178
Tara Pacific	2016	2018	Tropical Pacific–North Atlantic	Surface and subsurface	325
Tara Ocean	2009	2012	Global Ocean	Surface and 400 m	123
Tara Microbiome	2021	2022	Atlantic	Surface and water column	216
ACE	2016	2017	Southern Ocean	Surface and water column	135
MOBYDICK	2019		Southern Ocean	Water column	64
CLIM-EPARSES*	2019		Indian	Surface	790
OISO*	1998	2018	South Indian	Surface	24 950
DYFAMED	1998	2017	MedSea	Water column	2118
BOUSSOLE	2014	2019	MedSea	Surface and 10m	172
SOMLIT-Point-B	2007	2015	MedSea, coastal	Surface and 50 m	2397
ANTARES	2010	2016	MedSea	Water column	502
MOLA	2010	2013	MedSea, coastal	Water column	66
SOLEMIO	2016	2018	MedSea, coastal	Water column	212
MOOSE-GE	2010	2019	MedSea	Water column	1847
LATEX	2010		MedSea	Water column	51
CARBORHONE	2011	2012	MedSea	Water column	706
CASCADE	2011		MedSea	Water column	218
DEWEX	2013		MedSea	Water column	367
SOMBA	2014	2014	MedSea	Water column	203
AMOR-BFLUX	2015		MedSea, coastal	Water column	6
PEACETIME	2017	2017	MedSea	Water column	233
PERLE	2018	2021	MedSea	Water column	805

\* Measurements were taken at sea (surface measurement while underway).

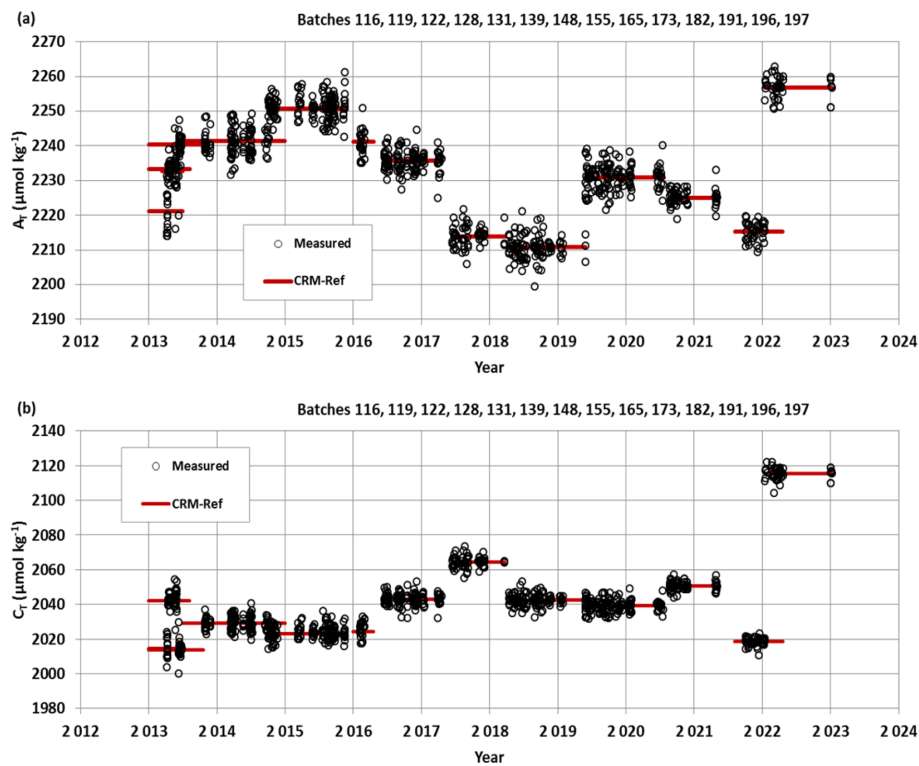
within  $0\text{--}3\ \mu\text{mol kg}^{-1}$ . Based on the CRM analyses and replicates for different projects, different regions and different periods, we estimated  $A_T$  and  $C_T$  data to be consistent with an accuracy of better than  $4\ \mu\text{mol kg}^{-1}$ .

### 3.3 Intercomparisons

Intercomparisons of measurements performed with different techniques help to evaluate the quality of the data and detect potential biases when merging different laboratories' data obtained from the same region during different periods. This is especially important to interpret long-term trends in  $A_T$  and  $C_T$  as well as for  $p\text{CO}_2$  and pH calculated with  $A_T\text{--}C_T$  pairs. For ocean acidification studies, this also refers to the



**Figure 1.** Locations of  $A_T$  and  $C_T$  data (1993–2022) in the Global Ocean and the western Mediterranean Sea (white box, inset) in the SNAPO-CO<sub>2</sub>-v1 dataset. The figure was produced with Ocean Data View (ODV; Schlitzer, 2018).



**Figure 2.**  $A_T$  (a) and  $C_T$  (b) analyses for different CRM batches measured in the 2013–2023 period. For these 965 analyses, the means (standard deviations) of the differences with the CRM reference were  $-0.1 (\pm 3.4) \mu\text{mol kg}^{-1}$  for  $A_T$  and  $0.1 (\pm 3.7) \mu\text{mol kg}^{-1}$  for  $C_T$ .

“climate goal”, for which an accuracy for  $A_T$  and  $C_T$  of better than  $\pm 2 \mu\text{mol kg}^{-1}$  is needed (Newton et al., 2015; Tilbrook et al., 2019). For the projects in this data synthesis, inter-laboratory comparisons were occasionally performed and are summarized below.

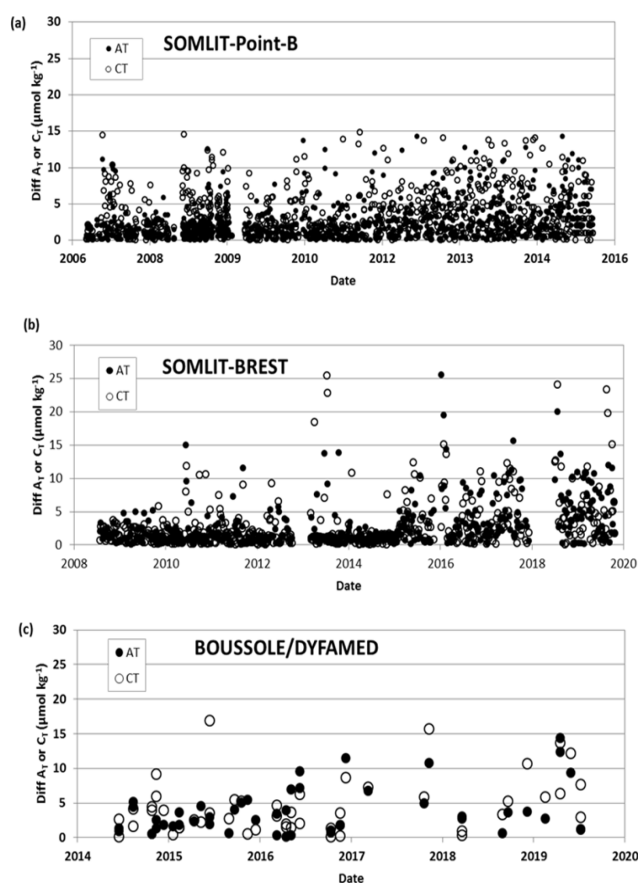
### 3.3.1 CHANNEL project

As part of the CHANNEL (2012–2015) time series in the western English Channel, Marrec et al. (2014) analyzed surface samples collected bimonthly from 2011 to 2013.  $A_T$  analyses were performed with a TA-ALK-2 system (Apollo SciTech), while  $C_T$  measurements were acquired with an AIRICA (Automated Infra Red Inorganic Carbon Analyzer) system (Marianda Inc.) Based on CRM analyses (batch

**Table 2.** Repeatability of the  $A_T$  and  $C_T$  analyses for cruises with duplicate analysis. The results are expressed as the standard deviation (SD) values of the analysis of replicated samples. Nb denotes the number of replicates for each time series or cruise (see Fig. 3 for the results of regular duplicates for three time series: SOMLIT-Point-B, SOMLIT-BREST and BOUSSOLE). For the 26 OISO cruises (1998–2018) and for simplicity, we list the mean repeatability obtained for all cruises.

Cruise	Nb	SD $A_T$ ( $\mu\text{mol kg}^{-1}$ )	SD $C_T$ ( $\mu\text{mol kg}^{-1}$ )	Reference
OUTPACE	12	3.64	3.68	Wagener et al. (2018a)
SOMBA	13	2.00	3.30	Keraghel et al. (2020)
SOMLIT-Point-B	786	2.63	3.10	Kapsenberg et al. (2017)
SOMLIT-BREST	446	3.34	3.67	Salt et al. (2016) and unpublished information
BOUSSOLE	48	3.47	4.02	Merlivat et al. (2018) and Golbol et al. (2020)
CLIM-EPARSEES	122	2.20	2.30	Lo Monaco et al. (2020, 2021)
OISO 1998–2018	1162	2.06	2.28	Metzl et al. (2006) and OISO Lines information <sup>a</sup>

<sup>a</sup> Metadata and data available at [https://www.nodc.noaa.gov/ocads/oceans/VOS\\_Program/OISO.html](https://www.nodc.noaa.gov/ocads/oceans/VOS_Program/OISO.html) (last access: 22 December 2023).



**Figure 3.** Results of duplicate  $A_T$  and  $C_T$  analyses from the following time series: (a) SOMLIT-Point-B in the coastal Mediterranean Sea (Kapsenberg et al., 2017), (b) SOMLIT-BREST in the Bay of Brest in the coastal Iroise Sea (Salt et al., 2016, and unpublished information) and (c) BOUSSOLE/DYFAMED in the Ligurian Sea (Merlivat et al., 2018; Golbol et al., 2020). The plots show differences in duplicates for both  $A_T$  (filled circles) and  $C_T$  (open circles). SD values of these duplicates are listed in Table 2.

no. 92), the accuracy was estimated to be  $\pm 3 \mu\text{mol kg}^{-1}$  for  $A_T$  and  $\pm 1.5 \mu\text{mol kg}^{-1}$  for  $C_T$  (Marrec et al., 2014). When undertaking a comparison with the samples measured at LOCEAN, Paris, for the year 2012, Marrec et al. (2014) concluded that the concentrations between the two methods were within  $\pm 2$  and  $\pm 3 \mu\text{mol kg}^{-1}$  for  $A_T$  and  $C_T$ , respectively. This is close to the aforementioned climate goal, offering confident results for long-term trend analysis of the carbonate system in this region.

### 3.3.2 SURATLANT project

In the framework of the SURATLANT project in the subpolar North Atlantic gyre, some samples collected at the same time (in 2005, 2006, 2010, 2015 and 2016) were also analyzed onshore for  $A_T$  and/or  $C_T$  by other laboratories using different techniques (e.g., coulometric method); these results have been summarized by Reverdin et al. (2018). For  $C_T$ , the mean (SD) differences between LOCEAN values and those from four other laboratories range between  $-0.7 (\pm 4.6)$  and  $-6.5 (\pm 3.4) \mu\text{mol kg}^{-1}$ , depending on the cruise. For  $A_T$ , the mean differences compared with two other laboratories range from  $-0.6 (\pm 4.1) \mu\text{mol kg}^{-1}$  to  $+2.3 (\pm 4.8) \mu\text{mol kg}^{-1}$ .

### 3.3.3 OVIDE project

During OVIDE cruises, which have been conducted since 2002 in the North Atlantic along a section from Greenland to Portugal (Lherminier et al., 2007; Mercier et al., 2015), samples have been taken (since 2006) to complement (for summer) the SURATLANT time series in the North Atlantic subpolar gyre (NASPG). The OVIDE samples at the surface and along the water column at a few stations were measured back at LOCEAN for  $A_T$  and  $C_T$  (Metzl et al., 2018). This enabled us to compare our data with the onboard measurements performed by the IIM (Instituto de Investigaciones Marinas) group in Vigo, Spain (e.g., Pérez et al., 2010, 2013, 2018; Vazquez-Rodriguez et al., 2012). The OVIDE data



have been regularly quality controlled as part of the CARINA and GLODAP data products (Velo et al., 2009; Key et al., 2010; Olsen et al., 2016, 2019, 2020). The results of inter-comparisons are gathered in Table 3. For OVIDE in 2006, we identified (for unknown reason) a large difference between our original  $A_T$  values and the  $A_T$  data qualified in GLODAP; thus, we corrected our  $A_T$  data by  $+7.2 \mu\text{mol kg}^{-1}$ . However, no correction was applied to  $C_T$  data. For other OVIDE cruises, differences in  $A_T$  values range between  $-4.5$  ( $\pm 4.11$ )  $\mu\text{mol kg}^{-1}$  and  $-0.05$  ( $\pm 3.43$ )  $\mu\text{mol kg}^{-1}$ , depending on the cruise (i.e.,  $A_T$  measured at LOCEAN was always slightly lower than onboard measurements). For  $C_T$ , we compared our onshore measurements with  $C_T$  values calculated with  $A_T$  and pH measured on board. Most of the mean  $C_T$  differences are slightly positive (i.e.,  $C_T$  measured at LOCEAN was always higher, except for 2010). Considering all errors associated with sampling, the transport of samples, the instrumentation, the data processing or the calculation of  $C_T$  using  $A_T$ -pH pairs (around  $8.8 \mu\text{mol kg}^{-1}$ ; Orr et al., 2018), the comparisons between LOCEAN and IIM data for OVIDE cruises are deemed acceptable, and large differences in both  $A_T$  and  $C_T$  ( $>4 \mu\text{mol kg}^{-1}$ ) are far from being systematic (Table 3). The data from SURATLANT and OVIDE can then be merged to complete the time series in the NASPG in summer and to better describe the seasonality of the oceanic carbonate system. For example, in 2010, when the North Atlantic Oscillation (NAO) was strongly negative, the SURATLANT data showed a rapid decrease in  $C_T$  concentrations in the NASPG between early-June and August (Fig. 4), with  $C_T$  concentrations in August being much lower than other years (Racapé et al., 2014). This led to a rapid drop in  $f\text{CO}_2$  in 2009–2010, such that the NASPG was a strong  $\text{CO}_2$  sink (Leseurre et al., 2020). The winter-to-summer seasonal decrease in  $C_T$  in 2010 in the northern NASPG was on average  $-77 \mu\text{mol kg}^{-1}$  (Fig. 4), much larger than in the climatology (range of  $-50$  to  $-55 \mu\text{mol kg}^{-1}$ ; Takahashi et al., 2014; Reverdin et al., 2018). The OVIDE data in late June in 2010 and SURATLANT in August 2010 confirmed this signal that was linked to pronounced primary productivity in that period (Fig. 4; Henson et al., 2013; Racapé et al., 2014; McKinley et al., 2018). Note that, for this period, no  $f\text{CO}_2$  observations were available in July–September 2010 in the SOCAT data product and that the  $A_T$  and  $C_T$  data presented here could be used to calculate  $f\text{CO}_2$  to complement the  $f\text{CO}_2$  dataset in this region, as was done for other periods (McKinley et al., 2011).

### 3.3.4 The river La Penzé

The comparisons described above concern the open-ocean region that has  $A_T$  and  $C_T$  concentrations in a range of concentrations close to the CRM references (used by the different laboratories). Another example of a comparison is presented here for samples obtained along a river and, thus, for waters with a low salinity and low  $A_T$  concentrations (river

La Penzé in northern Brittany). In 2019–2020,  $A_T$  was measured at SBR laboratory using a potentiometric method (with a Titrino 847 plus Metrohm) calibrated with CRM (batch no. 131) for a final accuracy of  $\pm 2.1 \mu\text{mol kg}^{-1}$  (Gac et al., 2020). Although the samples were measured with different techniques, the  $A_T$ -salinity relationships are very coherent for both datasets (Fig. 5). The regressions for each period for  $A_T$  (in  $\mu\text{mol kg}^{-1}$ ) are as follows:

$$\begin{aligned} - A_T &= 51.525 (\pm 0.944) S + 583.95 (\pm 19.94) (r^2 = 0.975) \text{ (for 78 samples in 2011);} \\ - A_T &= 54.022 (\pm 1.018) S + 450.23 (\pm 31.53) (r^2 = 0.976) \text{ (for 70 samples in 2019–2020).} \end{aligned}$$

Therefore, we added the  $A_T$  data measured in 2019–2020 to complete the synthesis for this location (river La Penzé).

### 3.4 Assigned flags for quality control

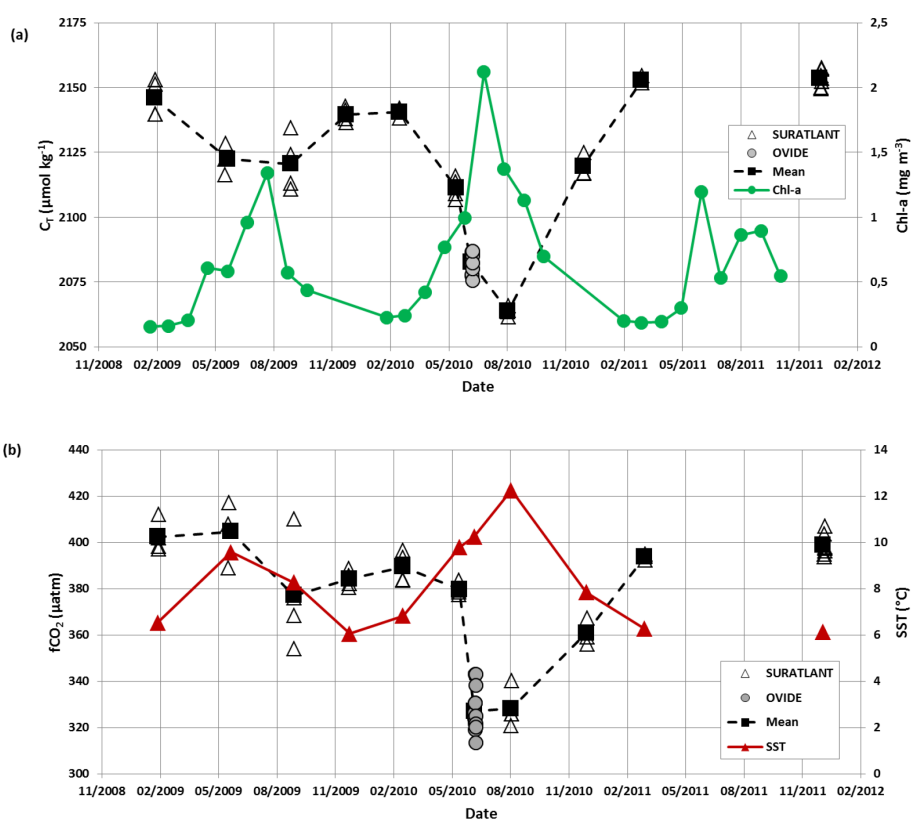
Identifying each data point with an appropriate flag is very convenient for selecting data (good, questionable or bad data). Here, following the WOCE program, we used four flags for each property (Flag 2 – good data, Flag 3 – questionable data, Flag 4 – bad data and Flag 9 – no data), as used in other data products such as SOCAT (Bakker et al., 2016) or GLODAP (Olsen et al., 2016, 2019, 2020; Lauvset et al., 2021). During data processing, we first assigned a flag for each  $A_T$  and  $C_T$  data point based on the standard error in the calculation of  $A_T$  and  $C_T$  concentrations (non-linear regression; Dickson et al., 2007). By default, if the standard deviation of the regression was  $>1 \mu\text{mol kg}^{-1}$ , we assigned Flag 3 (questionable), although the data could be acceptable and then used for interpretations. Flag 3 was also assigned when salinity was doubtful or when differences between duplicates were large (e.g.,  $\pm 20 \mu\text{mol kg}^{-1}$ ). Flag 4 (bad or certainly bad) was assigned when clear anomalies were detected for unknown reasons (e.g., a sample probably not fixed with  $\text{HgCl}_2$ ). A secondary quality control was performed by the principal investigator of each project based on data inspection, duplicates, the  $A_T$ -salinity relationship, or the mean observations in deep layers where large variability in  $A_T$  and  $C_T$  is unlikely to occur from year to year. An example presents all data from the MOOSE-GE cruises conducted in 2010–2019 in the Mediterranean Sea (Coppola et al., 2020; Testor et al., 2010) where clear outliers have been identified (Fig. S3). For the 10 MOOSE-GE cruises and a total of 1847  $A_T$  and  $C_T$  analyses, 26 data points were flagged as bad (Flag 4), 139 data points for  $A_T$  and 141 data points for  $C_T$  were listed as questionable (Flag 3), and 1682 data points for  $A_T$  and 1680 data points for  $C_T$  were considered to be good data (Flag 2, i.e., more than 90%). A similar control process was performed for each project.

The synthesis of various cruises in the same region and period also offers verification and secondary control of the data. For example, several cruises were conducted in the Mediterranean Sea in 2014 (MOOSE-GE, SOMBA, ANTARES and

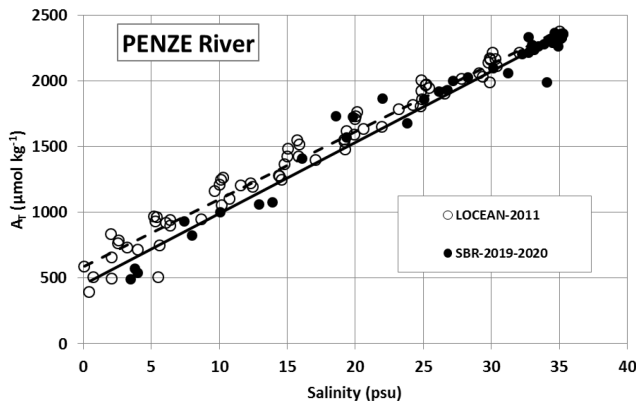
**Table 3.** Comparisons of  $A_T$  and  $C_T$  samples measured back at LOCEAN with onboard measurements by IIM laboratory (Fiz Pérez, Vigo, Spain) for OVIDE cruises in the North Atlantic. Nb denotes the number of samples and ND denotes no data. The results listed indicate the means and standard deviations of the differences (LOCEAN – IIM). For  $A_T$ , IIM values were measured on board. For  $C_T$ , IIM values were calculated from  $A_T$  and pH, both measured on board. Here, the IIM data were quality controlled and taken from the GLODAP data products (Olsen et al., 2016, 2019).

Cruise and year	Nb $A_T$	$A_T$ (LOCEAN) – $A_T$ (IIM) $\mu\text{mol kg}^{-1}$	Nb $C_T$	$C_T$ (LOCEAN) – $C_T$ (IIM) $\mu\text{mol kg}^{-1}$
OVIDE 2006	14	–2.0 ( $\pm 5.9$ )*	14	1.1 ( $\pm 2.5$ )
OVIDE 2008	29	–4.5 ( $\pm 4.1$ )	29	3.8 ( $\pm 3.1$ )
OVIDE 2010	41	–2.0 ( $\pm 2.3$ )	41	–2.4 ( $\pm 3.3$ )
OVIDE 2012	37	–0.1 ( $\pm 8.8$ )	ND	ND
GEOVIDE 2014	57	–0.1 ( $\pm 3.4$ )	54	2.4 ( $\pm 7.9$ )

\* For the OVIDE 2006 cruise, the original difference in  $A_T$  was  $-9.0 (\pm 5.8) \mu\text{mol kg}^{-1}$ . LOCEAN  $A_T$  data were corrected by  $+7.2 \mu\text{mol kg}^{-1}$  based on the mean concentrations in deep layers, whereas no corrections were applied for  $A_T$  and  $C_T$  for other cruises.



**Figure 4.** (a) Time series of  $C_T$  concentrations ( $\mu\text{mol kg}^{-1}$ ) for 2009–2011 in surface waters in the North Atlantic subpolar gyre (zone  $59^{\circ}\text{N}$ ,  $33^{\circ}\text{W}$ ) based on SURATLANT (open triangles) and OVIDE 2010 (gray circles) data. In 2009, SURATLANT data were available in February, June, September and December; in 2010, data were available in March, June, August and December; and in 2011, data were only available for March and December. The OVIDE data in late June in 2010 completed the temporal cycle and confirmed the strong seasonal signal and low  $C_T$  concentrations in summer 2010 not seen in 2009 (nor in 2011, as there were no data in summer). The mean observations for each period describe the  $C_T$  seasonal cycles in 2009 and 2010 (black squares, dashed line). The monthly surface chlorophyll-*a* (Chl-*a*) concentrations ( $\text{mg m}^{-3}$ ) averaged in the same region based on MODIS are also shown (green dots and line), highlighting the high productivity during the summer 2010. Chl-*a* monthly data were extracted from MODIS (Giovanni, NASA, last access: 3 May 2019). (b) Time series of  $f\text{CO}_2$  ( $\mu\text{atm}$ ) for the same cruises (same symbols) were calculated with  $A_T$  and  $C_T$  and using the  $K_1$  and  $K_2$  constants from Lueker et al. (2000). The mean sea-surface temperature (SST,  $^{\circ}\text{C}$ ) is also indicated (red triangles). In June 2010, oceanic  $f\text{CO}_2$  decreased by  $53 \mu\text{atm}$  in 2 weeks.



**Figure 5.** Total alkalinity ( $A_T$ ) versus salinity for samples measured in 2011 and 2019 in the river La Penzé, northern Brittany (Gac et al., 2020).  $A_T$  samples were measured at LOCEAN in 2011 (open circles, dashed line) and at SBR laboratory (Roscoff) in 2019 (filled circles, black line).

DYFAMED). The mean values of  $C_T$  and  $A_T$  in the deep layers (>1800 m) for each cruise confirmed the coherence of the data (Table 4). This enabled us to merge the different datasets in order to interpret the temporal trends and processes driving the  $\text{CO}_2$  cycle (Coppola et al., 2019, 2020) or to train and validate a regional neural network to reconstruct the carbonate system (e.g., CANYON-MED; Fourrier et al., 2020, 2022).

The total number of data for the Global Ocean and the Mediterranean Sea are gathered in Table 5 with the corresponding flags for each property. Overall, the synthesis includes more than 94 % of good data for both  $A_T$  and  $C_T$ . About 5 % of data are questionable and 2 % are likely bad. Overall, we believe that all data (with Flag 2) in this synthesis have an accuracy of better than  $4 \mu\text{mol kg}^{-1}$  for both  $A_T$  and  $C_T$ , which is the same as for quality-controlled data in GLODAP (Olsen et al., 2020; Lauvset et al., 2021). The uncertainty ranges between the climate goal ( $2 \mu\text{mol kg}^{-1}$ ) and the “weather goal” ( $10 \mu\text{mol kg}^{-1}$ ) for ocean acidification studies (Newton et al., 2015; Tilbrook et al., 2019). This accuracy is also relevant to validate or constrain data-based methods that reconstruct  $A_T$  and  $C_T$  fields with an error of around  $10\text{--}15 \mu\text{mol kg}^{-1}$  for both properties (Bittig et al., 2018; Broullón et al., 2019, 2020; Fourrier et al., 2020).

### 3.5 Using $A_T$ and $C_T$ to calculate $f\text{CO}_2$ and pH for comparison with $f\text{CO}_2$ and pH measurements

For some projects, the  $A_T$  and  $C_T$  data presented in this synthesis were used to calibrate or validate in situ  $f\text{CO}_2$  sensors (Bozec et al., 2011; Marrec et al., 2014; Merlivat et al., 2018). The  $A_T$  and  $C_T$  data were also used to calculate  $f\text{CO}_2$  and to derive associated air–sea  $\text{CO}_2$  fluxes, especially during periods when no direct  $f\text{CO}_2$  measurements were available (e.g., in the North Atlantic; Fig. 4; Watson et al., 2009;

McKinley et al., 2011). For example, Marrec et al. (2014) successfully used the calculated  $p\text{CO}_2$  (with  $A_T\text{--}C_T$  pairs) to adjust the drift of the  $p\text{CO}_2$  data recorded with a CONTROS HydroC  $\text{CO}_2$  FT sensor mounted on a FerryBox for regularly sampling the western English Channel (CHANNEL project). Here, we show the results for the 2012–2014 period (Fig. 6). In this region, the total alkalinity is relatively constant over time: the average  $A_T$  for 528 samples from different seasons and years is  $2334.4 (\pm 7.2) \mu\text{mol kg}^{-1}$ . Conversely, the  $C_T$  concentrations show distinctive seasonality, with higher concentrations in winter and lower concentrations in summer when biological activity is pronounced (Marrec et al., 2013, 2014; Kitidis et al., 2019). This controls the seasonal  $p\text{CO}_2$  distribution revealed each year in both the measured and calculated  $p\text{CO}_2$  (Fig. 6). For 528 co-located samples, the mean difference between calculated and measured  $p\text{CO}_2$  is  $-1.9 (\pm 11.9) \mu\text{atm}$ , with no distinct differences depending on the season and year.

In the Ligurian Sea, following the first high-frequency in situ  $f\text{CO}_2$  measurements in 1995–1997 at the DYFAMED time series’ station (Hood and Merlivat, 2001), a new CARIOCA  $f\text{CO}_2$  sensor was deployed at that location in 2013 (BOUSSOLE project; Merlivat et al., 2018). The CARIOCA sensor was calibrated with regular  $A_T$  and  $C_T$  analyses performed at LOCEAN. Based on these data, the mean difference between CARIOCA  $f\text{CO}_2$  measurements and calculated  $f\text{CO}_2$  data was estimated to be around  $\pm 4.4 \mu\text{atm}$  for 2013–2015, i.e., of the same order as the precision of the CARIOCA sensor ( $\pm 5 \mu\text{atm}$ ; Merlivat et al., 2018). Here, we extend the results for the 2013–2018 period (Golbol et al., 2020; data also in SOCAT version v2021 in Bakker et al., 2016) and compared the CARIOCA  $f\text{CO}_2$  time series with  $A_T$  and  $C_T$  data from different cruises (BOUSSOLE, DYFAMED and MOOSE-GE) selected in the 0–20 m layer at that location (Fig. S4). For 67 co-located samples from different seasons and years, the mean difference between calculated and measured  $f\text{CO}_2$  ( $f\text{CO}_{2\text{cal}} - f\text{CO}_{2\text{mes}}$ ) was  $-3.7 (\pm 10.8) \mu\text{atm}$  where  $f\text{CO}_2$  was calculated from  $A_T\text{--}C_T$  pairs using the constant from Lueker et al. (2000). At the location of the BOUSSOLE mooring in the Ligurian Sea (shown in Fig. S4), the alkalinity is relatively constant over the 2013–2018 period, with an average concentration of  $2569.8 (\pm 13.2) \mu\text{mol kg}^{-1}$ . In contrast,  $C_T$  concentrations show a clear seasonality, decreasing by around  $50 \mu\text{mol kg}^{-1}$  from winter to late summer, driving the large seasonal cycle of  $f\text{CO}_2$  (range  $80 \mu\text{atm}$ ) revealed in both measured and calculated values (here,  $f\text{CO}_2$  is normalized at  $13^\circ\text{C}$ ; Fig. S4). In addition to calibration purposes, a regional  $A_T\text{--}salinity$  relationship was derived from the  $A_T$  data measured at that location and successfully used to construct time series of  $C_T$  and pH calculated from the high-frequency CARIOCA  $f\text{CO}_2$  data to investigate and interpret the long-term change in  $f\text{CO}_2$  and acidification in the Ligurian Sea (Merlivat et al., 2018; Coppola et al., 2020).

**Table 4.** Mean observations in the deep layers (>1800 m) of the western Mediterranean Sea for different cruises conducted in 2014. Results for deep layers (>1800 m) for the DEWEX cruise in 2013 and the PEACETIME cruise in 2017 in the same region are also listed.  $N-A_T$  and  $N-C_T$  are  $A_T$  and  $C_T$  normalized at a salinity of 38, respectively. Nb denotes the number of data (with Flag 2). Standard deviations are shown in parentheses. References for these cruises are listed in the Supplement.

Cruise	Period	Nb	Potential temperature (°C)	Salinity (PSU)*	$N-A_T$ ( $\mu\text{mol kg}^{-1}$ )	$N-C_T$ ( $\mu\text{mol kg}^{-1}$ )
All cruises	February–December 2014	76	12.905 (0.007)	38.486 (0.005)	2562.9 (5.3)	2303.7 (4.7)
ANTARES	February–November 2014	14	12.913 (0.004)	38.488 (0.006)	2564.0 (3.8)	2301.9 (3.5)
DYFAMED	March–December 2014	9	12.905 (0.0016)	38.487 (0.004)	2560.1 (5.0)	2304.3 (6.8)
MOOSE-GE	July 2014	21	12.909 (0.004)	38.487 (0.005)	2565.6 (4.6)	2303.5 (4.1)
SOMBA	August–September 2014	32	12.899 (0.005)	38.483 (0.005)	2561.5 (5.6)	2304.6 (4.8)
DEWEX	February–April 2013	44	12.903 (0.010)	38.588 (0.006)	2556.0 (4.3)	2294.0 (5.7)
PEACETIME	May–June 2017	7	12.904 (0.002)	38.486 (0.003)	2567.2 (10.6)	2308.1 (8.9)

\* PSU represents practical salinity units.

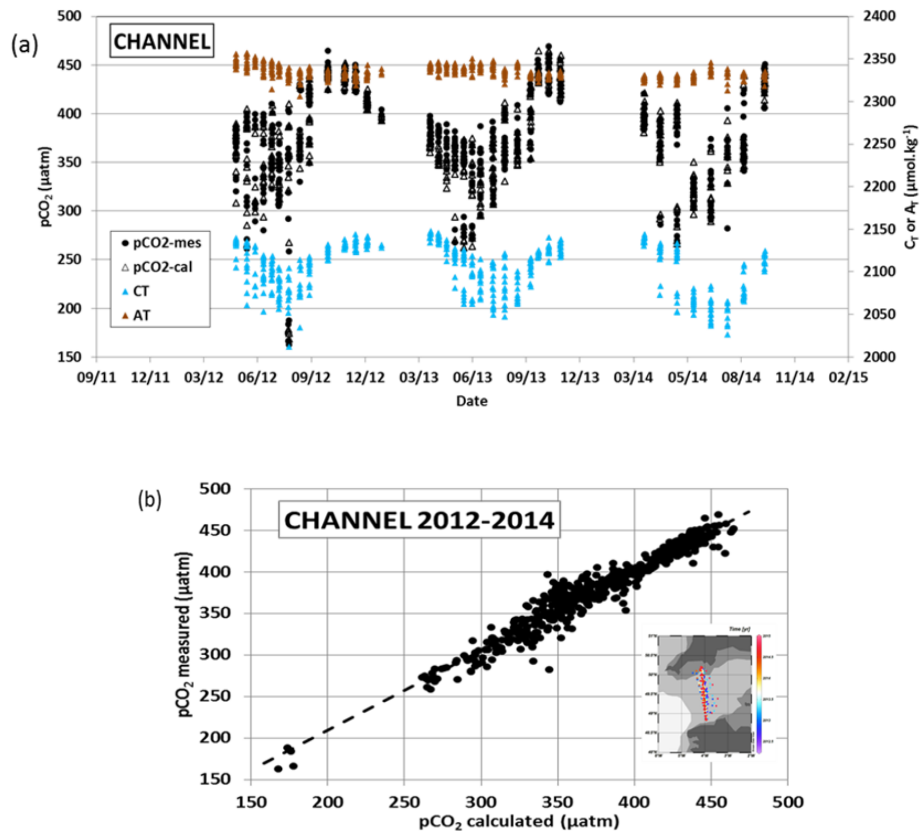
**Table 5.** Number of temperature, salinity,  $A_T$  and  $C_T$  data in the synthesis identified for flags 2, 3, 4 and 9. The data are given for the full Global Ocean and for the Mediterranean Sea dataset. The last column is the percentage of Flag 2 (good).

	Flag 2	Flag 3	Flag 4	Flag 9	% Flag 2
Global Ocean					
Temperature	43 538	410	0	478	99.07
Salinity	44 033	319	2	71	99.28
$A_T$	39 331	2144	1165	1787	92.24
$C_T$	39 921	2091	1148	1279	92.50
Mediterranean Sea					
Temperature	9843	1	0	65	99.99
Salinity	9879	8	2	20	99.99
$A_T$	8853	425	411	220	91.37
$C_T$	8854	451	389	211	91.33

A CARIOCA sensor was also deployed in 2003 near the SOMLIT-BREST time series' site in the Bay of Brest (Bozec et al., 2011; Salt et al., 2016). As for BOUSSOLE in the Ligurian Sea, samples collected for  $A_T$  and  $C_T$  were used for validation of the  $p\text{CO}_2$  recorded by the CARIOCA sensor, and the comparison with calculated  $p\text{CO}_2$  showed good agreement, i.e.,  $p\text{CO}_{2\text{cal}} = 0.98 \times p\text{CO}_{2\text{mes}} + 7 \mu\text{atm}$  (Bozec et al., 2011). CARIOCA sensors were also deployed on moorings in the tropical Atlantic (PIRATA project; e.g., Lefèvre et al., 2008, 2016; Parard et al., 2010). With the discrete  $A_T$  and  $C_T$  data included in this synthesis (EGEE

and PIRATA-FR cruises), the  $f\text{CO}_2$  data from the CARIOCA sensor associated with an adapted  $A_T$ –salinity relationship were used to derive pH (Lefèvre et al., 2016) or  $C_T$  time series to evaluate net community production in the eastern tropical Atlantic (Parard et al., 2010; Lefèvre and Merlivat, 2012).

Although this is not a direct instrumental intercomparison, differences between  $p\text{CO}_2$  (or  $f\text{CO}_2$ ) calculated using  $A_T$ – $C_T$  pairs with direct  $p\text{CO}_2$  measurements provide a glimpse into the quality of  $A_T$  and  $C_T$  data in this synthesis, given the uncertainty attached to the  $p\text{CO}_2$  or pH calculations (Orr



**Figure 6.** Panel (a) presents time series of  $A_T$  (brown triangles, right y axis),  $C_T$  (blue triangles, right y axis), calculated  $p\text{CO}_2$  (open triangles, left y axis) and measured  $p\text{CO}_2$  (filled circles) in the western English Channel in 2012–2014 (Marrec et al., 2014). Panel (b) shows measured  $p\text{CO}_2$  versus calculated  $p\text{CO}_2$  for the same samples. The mean difference ( $p\text{CO}_{2\text{cal}} - p\text{CO}_{2\text{mes}}$ ) for 528 samples is  $-1.9 (\pm 11.9) \mu\text{atm}$ . Data are from Marrec and Bozec (2016a, b, 2017). Localization of the samples is shown in the inset map in panel (b), which was produced with ODV (Schlitzer, 2018).

et al., 2015). For example, in the framework of the SURATLANT project in the North Atlantic, calculated  $f\text{CO}_2$  data were compared with co-located  $f\text{CO}_2$  measurements for different seasons and years (Fig. S5). The mean differences ( $f\text{CO}_{2\text{cal}} - f\text{CO}_{2\text{mes}}$ ) ranged between  $-4.3 (\pm 12.9) \mu\text{atm}$  (2004–2007, 74 co-located samples) and  $-3.0 (\pm 12.1) \mu\text{atm}$  (2014–2015, 98 co-located samples). The differences are almost the same for different years (and seasons) and are, thus, attributed to method uncertainties (including sampling time, measurement errors and data processing). Based on these comparisons and the consistency between data, we are confident that the  $A_T$  and  $C_T$  data presented in this synthesis could be used to calculate  $f\text{CO}_2$  (and pH) and to interpret temporal changes and drivers of these parameters as well as to estimate air–sea CO<sub>2</sub> fluxes in the North Atlantic (e.g., Corbière et al., 2007; Schuster et al., 2009, 2013; Watson et al., 2009; Metzler et al., 2010; McKinley et al., 2011; Reverdin et al., 2018; Kitidis et al., 2019; Leseurre et al., 2020).

The  $A_T$  and  $C_T$  data in this synthesis have also been successfully used for  $f\text{CO}_2$  and air–sea CO<sub>2</sub> flux calculations in other regions: the tropical Atlantic (Koffi et al., 2010), the

tropical Pacific (Moutin et al., 2018; Wagener et al., 2018a), the Solomon Sea (Ganachaud et al., 2017), or the Mediterranean Sea and coastal zones (De Carlo et al., 2013; Marrec et al., 2015; Kapsenberg et al., 2017; Coppola et al., 2020; Keraghel et al., 2020; Wimart-Rousseau et al., 2020; Gattuso et al., 2023).

In addition,  $A_T$  and  $C_T$  data at the surface and in the water column are also relevant to calculate pH and evaluate its rate of change in the context of addressing the topic of ocean acidification in different regions (Kapsenberg et al., 2017; Ganachaud et al., 2017; Wagener et al., 2018a; Coppola et al., 2020; Leseurre et al., 2020; Lo Monaco et al., 2021). At the ECOSCOPIA time series’ station in the Bay of Brest (Fleury et al., 2023; Petton et al., 2023), pH values calculated with  $A_T$  and  $C_T$  data were compared with direct pH measurements (Fig. S6). In 2017–2019, pH (at a standard temperature of 25 °C, hereafter pH-25C) was always lower than 8 and presented a large seasonal signal of 0.3 (high pH values in spring but low values in winter). The mean difference between calculated and measured pH-25C for 46 samples was equal to  $+0.013 (\pm 0.010)$ , which is in the range of the pH uncertainty

evaluated by error propagation when calculated from  $A_T$ – $C_T$  pairs (an  $A_T$  and  $C_T$  error of  $\pm 3 \mu\text{mol kg}^{-1}$  leads to a pH error of  $\pm 0.0144$ ). Part of these  $A_T$  and  $C_T$  data used to calculate pH also helped to interpret the response of marine species to acidification, e.g., pteropodes or coccolithophores (*Emiliania huxleyi*), in the Mediterranean Sea (Howes et al., 2015, 2017; Meier et al., 2014) or in the Southern Ocean (Beaufort et al., 2011). The  $A_T$  and  $C_T$  data also supported environmental analysis in coral reef ecosystems in the tropical Pacific (Tara expedition; Douville et al., 2022; Lombard et al., 2023; Canesi et al., 2023).

#### 4 Global distribution and relationships from $A_T$ and $C_T$ based on the SNAPO-CO<sub>2</sub> dataset

The surface distribution in the Global Ocean based on the SNAPO-CO<sub>2</sub> dataset is presented in Fig. 7 for  $A_T$  and  $C_T$ . In the open ocean, high  $A_T$  concentrations are identified in the subtropics in all basins (Jiang et al., 2014; Takahashi et al., 2014), with the highest concentrations of up to  $2484 \mu\text{mol kg}^{-1}$  found in the central North Atlantic (STRASSE cruise in August 2012;  $26^\circ \text{N}$ ,  $36^\circ \text{W}$ ). At the surface and at depth, the  $A_T$ –salinity and  $A_T$ – $C_T$  relationships are clearly identified and structured at the regional scale (Fig. 8).

In the eastern tropical Atlantic (ETA), where the Congo River impacts the salinity field (Vangriesheim et al., 2009),  $A_T$  concentrations range between  $2100$  and  $2400 \mu\text{mol kg}^{-1}$ . The regional  $A_T$ –salinity relationship in the ETA based on data from the EGEE cruises in 2005–2007 (Koffi et al., 2010) is robust and has been validated with more recent measurements from PIRATA-FR cruises in 2010–2019 (Lefèvre et al., 2021). The strong  $A_T$ –salinity relationship in the ETA was also recognized using data from the Tara Microbiome cruise in May–July 2022 (Fig. S7). Low salinity ( $<30$ ) and low  $A_T$  ( $1700$ – $2200 \mu\text{mol kg}^{-1}$ ) are also observed in the western tropical Atlantic near the Amazon River plume. The  $A_T$ –salinity relationships in both river plume regions are very similar (Fig. S7).

For  $C_T$ , the lowest concentrations were observed in the coastal regions of the tropical Atlantic, on the eastern side of the Gulf of Guinea ( $C_T = 1390 \mu\text{mol kg}^{-1}$ , BIOZAIRE cruise in 2003;  $6^\circ \text{S}$ ,  $11^\circ \text{E}$ ; Vangriesheim et al., 2009) and on the western side of the Gulf of Guinea in the coastal zone off French Guyana ( $C_T = 1512 \mu\text{mol kg}^{-1}$ , PLUMAND cruise in 2007;  $5^\circ \text{N}$ ,  $51^\circ \text{W}$ ; Lefèvre et al., 2010). Such low  $C_T$  concentrations were also observed in the Amazon River plume (around  $5^\circ \text{N}$ ,  $51^\circ \text{W}$ ) during the recent EUREC4A-OA cruise in 2020 and the Tara Microbiome cruise in 2021 ( $C_T = 1451 \mu\text{mol kg}^{-1}$ ) leading to low oceanic  $f\text{CO}_2$  ( $<350 \mu\text{atm}$ ) and a  $\text{CO}_2$  sink in this region (Olivier et al., 2022).

The high  $C_T$  concentrations were mainly observed in the Southern Ocean (OISO and Antarctic Circumpolar Expedi-

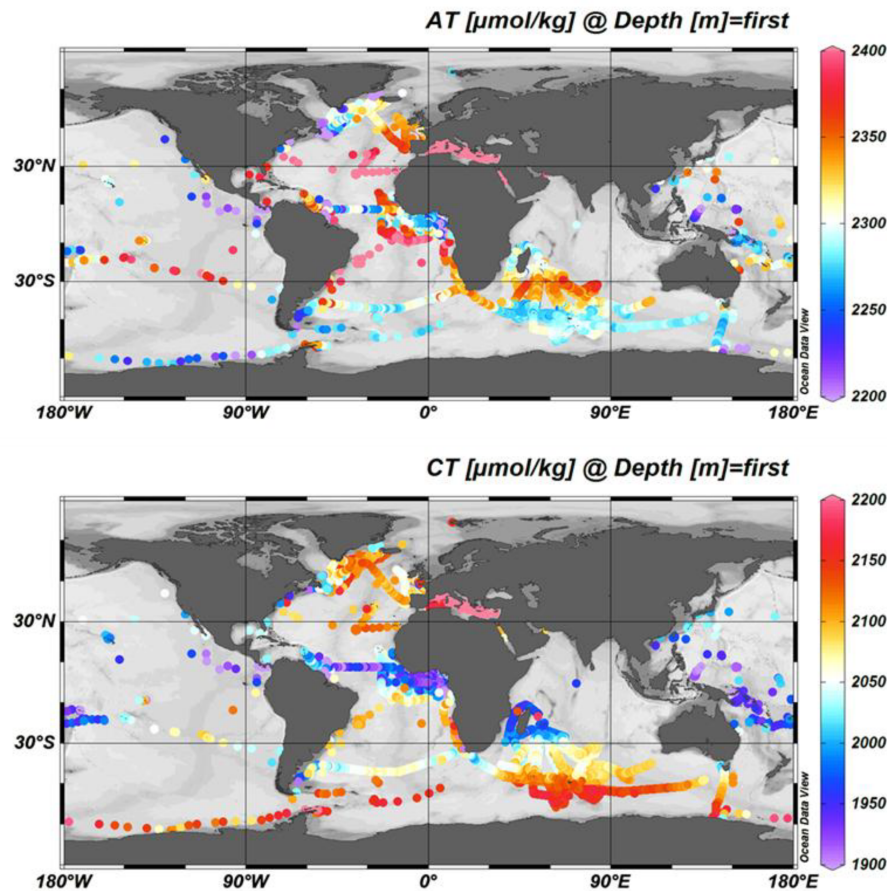
tion, ACE, cruises), south of the polar front around  $50^\circ \text{S}$ , linked to the upwelling of  $C_T$ -rich deep water (Fig. 7, Metzl et al., 2006; Wu et al., 2019; Chen et al., 2022). This leads to a high  $C_T/A_T$  ratio and a high Revelle factor in the Southern Ocean (Fig. 9; Fassbender et al., 2017). The high  $C_T$  content and low temperature in the Southern Ocean also lead to a low calcite and aragonite saturation state ( $\Omega$ ) (Takahashi et al., 2014; Jiang et al., 2015). We calculate  $\Omega$  from  $A_T$  and  $C_T$  data at in situ temperature, salinity and pressure. At present, the surface ocean is not undersaturated with regard to aragonite (Fig. 10); however, undersaturation levels ( $\Omega\text{-Ar} < 1$ ) were found at around 500 m in the Southern Ocean (ACE cruise in 2017 and MODYDICK cruise in 2018), 1000 m in the tropical Pacific (PANDORA 2012 and OUTPACE 2015 cruises) and 2200 m in the North Atlantic (OVIDE 2012 and 2014 cruises; see also Turk et al., 2017) (Fig. 10). Samples at 400 m from the Tara Oceans cruise in 2009–2012 also indicated aragonite undersaturation in the equatorial Atlantic, equatorial Pacific and off South America ( $34^\circ \text{S}$ ,  $73^\circ \text{W}$ ; Chile) associated with equatorial or eastern boundary upwelling systems (Feely et al., 2012; Lauvset et al., 2020).

At the surface,  $\Omega\text{-Ar} > 3$  is found in the latitudinal band between  $45^\circ \text{S}$  and  $54^\circ \text{N}$ , whereas  $\Omega\text{-Ar} < 3$  below the critical threshold of  $\Omega\text{-Ar} = 3.25$  that represents a limit for the distribution of tropical coral reefs (Hoegh-Guldberg et al., 2007) is observed at very few locations in the tropics.

Compared with the open ocean,  $A_T$  concentrations are much higher in the Mediterranean Sea (Copin-Montégut, 1993; Schneider et al., 2007; Álvarez et al., 2023), with values up to  $2600 \mu\text{mol kg}^{-1}$  (Fig. 8). The  $A_T$  and  $C_T$  data obtained in 1998–2019 show, on average, a clear contrast between the northern and southern regions of the western Mediterranean Sea (Fig. 11a, b), with a higher concentration in the Ligurian Sea and the Gulf of Lion (Gemayel et al., 2015). However, the basin-scale average distribution view smoothed the mesoscale signals recognized in the Mediterranean Sea (e.g., Bosse et al., 2017; Petrenko et al., 2017). In the Gulf of Lion, the synthesis of 11 cruises conducted from May 2010 to June 2011 (CARBORHONE, CASCADE, LATEX, MOLA and MOOSE-GE) highlights the contrasting distributions of  $A_T$  and  $C_T$  in the coastal zones and offshore (Fig. 11c, d). The averaging of all data for 1998–2019 also smoothed the seasonal signal and the interannual variability described below.

#### 5 Temporal variations in $A_T$ and $C_T$ : examples from the SNAPO-CO<sub>2</sub> dataset

Time series' stations, such as BATS, ESTOC and HOT, in the subtropics and stations in the Irminger Sea or in the Iceland Sea are the only way to detect the long-term change in the ocean carbonate system at the surface and in the water column (Bates et al., 2014). These important time series help to



**Figure 7.** Distribution of  $A_T$  (top) and  $C_T$  (bottom) concentrations ( $\mu\text{mol kg}^{-1}$ ) in surface waters (0–10 m). Only data with Flag 2 are presented in these figures. The figure was produced with ODV (Schlitzer, 2018).

understand driving processes (e.g., Hagens and Middelburg, 2016) and are often used to validate the reconstructed  $p\text{CO}_2$ ,  $A_T$ ,  $C_T$  or pH fields (e.g., Rödenbeck et al., 2013; Broullón et al., 2019, 2020; Keppler et al., 2020; Gregor and Gruber, 2021; Ma et al., 2023).

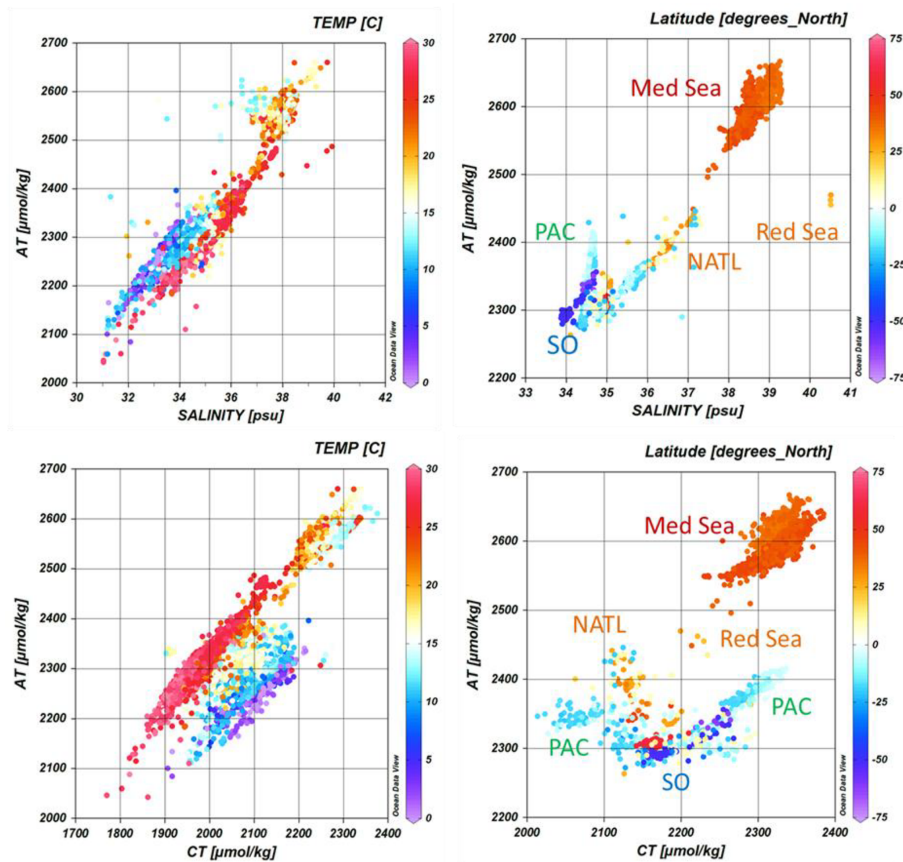
Here, we show examples of the temporal surface variations at locations where data were obtained for more than 10 years (Fig. 12). Thus, we selected the following contrasting regions: the North Atlantic subpolar gyre (NASPG; around  $60^\circ\text{N}$ ,  $30^\circ\text{W}$ ; 1993–2018 period), the equatorial Atlantic (at  $2^\circ\text{N}$ – $2^\circ\text{S}$ ,  $12^\circ\text{W}$ – $8^\circ\text{W}$ ; 2005–2017 period), the Indian Ocean subtropical sector ( $26$ – $35^\circ\text{S}$ ,  $50$ – $56^\circ\text{E}$ ; 1998–2018 period), the high-latitude Indian Ocean ( $54$ – $60^\circ\text{S}$ ,  $60$ – $70^\circ\text{E}$ ; 1998–2018 period), the Ligurian Sea (around DYFAMED station;  $43.5$ – $42.5^\circ\text{N}$ ,  $5.5$ – $9^\circ\text{E}$ ; 1998–2019 period) and times series' stations in the coastal zones off Brittany (2008–2019 period).

In the six regions, there was a progressive warming that was most clearly detected in the Mediterranean Sea (e.g., Nykjaer, 2009). From 1998 to 2019, the warming in the Ligurian Sea was  $+0.1208^\circ\text{C yr}^{-1}$  ( $\pm 0.0227^\circ\text{C yr}^{-1}$ ) (Fig. 12). In the equatorial Atlantic, the apparent rapid in-

crease in temperature of  $+0.48^\circ\text{C yr}^{-1}$  ( $\pm 0.04^\circ\text{C yr}^{-1}$ ) in 2005–2017 from the selected data indicated a change in the water masses and circulation. The colder sea surface in 2005 was associated with the so-called Atlantic cold tongue (ACT), which was one of the most intense ACTs since 1982 (Caniaux et al., 2011). The ACT also led to significant changes in oceanic  $f\text{CO}_2$  and air–sea  $\text{CO}_2$  fluxes (Parard et al., 2010; Koseki et al., 2023) and explained the high  $C_T$  concentrations observed in 2005 in this region (Fig. 12, Koffi et al., 2010).

Total alkalinity presents rather homogenous concentrations in the NASPG and the southern Indian Ocean. Inter-annual variability in  $A_T$  is pronounced in the equatorial Atlantic, ranging between 2245 and  $2378\ \mu\text{mol kg}^{-1}$ . This is mainly related to salinity, as normalized  $A_T$  values ( $N\text{-}A_T$ , for a salinity of 35) do not show such interannual variability (mean  $N\text{-}A_T = 2295.7 \pm 4.6\ \mu\text{mol kg}^{-1}$  and  $n = 67$  for 2005–2017; not shown). In the coastal zones off Brittany, the  $A_T$  is also highly variable (Salt et al., 2016; Gac et al., 2021), ranging between 2150 and  $2386\ \mu\text{mol kg}^{-1}$  (Fig. 12).

An interesting signal is the progressive increase in  $A_T$  in the Mediterranean Sea. The positive  $A_T$  trend of  $+0.53$



**Figure 8.** Relationships between  $A_T$  and salinity (upper panels) and between  $A_T$  and  $C_T$  (lower panels) for samples in surface waters (0–10 m and sea surface salinity (SSS) > 31) (left) and in the water column below 100 m (right). Only data with Flag 2 are presented. The color scales correspond to the temperature (left) or the latitude (right). The following data locations are identified: Mediterranean Sea (Med Sea), Red Sea, tropical Pacific (PAC), North Atlantic (NATL) and Southern Ocean (SO). The figure was produced with ODV (Schlitzer, 2018).

( $\pm 0.11$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  ( $n = 538$ ) in 1998–2019 in the off-shore region was also observed at the SOMLIT-Point-B coastal station in 2007–2015, although with a faster increase of  $+2.08$  ( $\pm 0.19$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  (Kapsenberg et al., 2017). Close to the DYFAMED site, at SOMLIT-Point-B station, the  $A_T$  trend was not linked to temporal salinity changes, as a positive N- $A_T$  trend was also reported,  $+0.52$  ( $\pm 0.07$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  (not shown). Based on data from the PERLE cruises in 2018–2021, a significant increase in  $A_T$  was also identified in the eastern Mediterranean Sea (Wimart-Rousseau et al., 2021). Along with the increase in  $C_T$  and the warming, the  $A_T$  increase would impact on the  $f\text{CO}_2$ , the air–sea  $\text{CO}_2$  fluxes and pH temporal changes (Merlivat et al., 2018). Processes explaining the  $A_T$  increase in the Mediterranean Sea are still unexplained and deserve further investigation (Coppola et al., 2019).

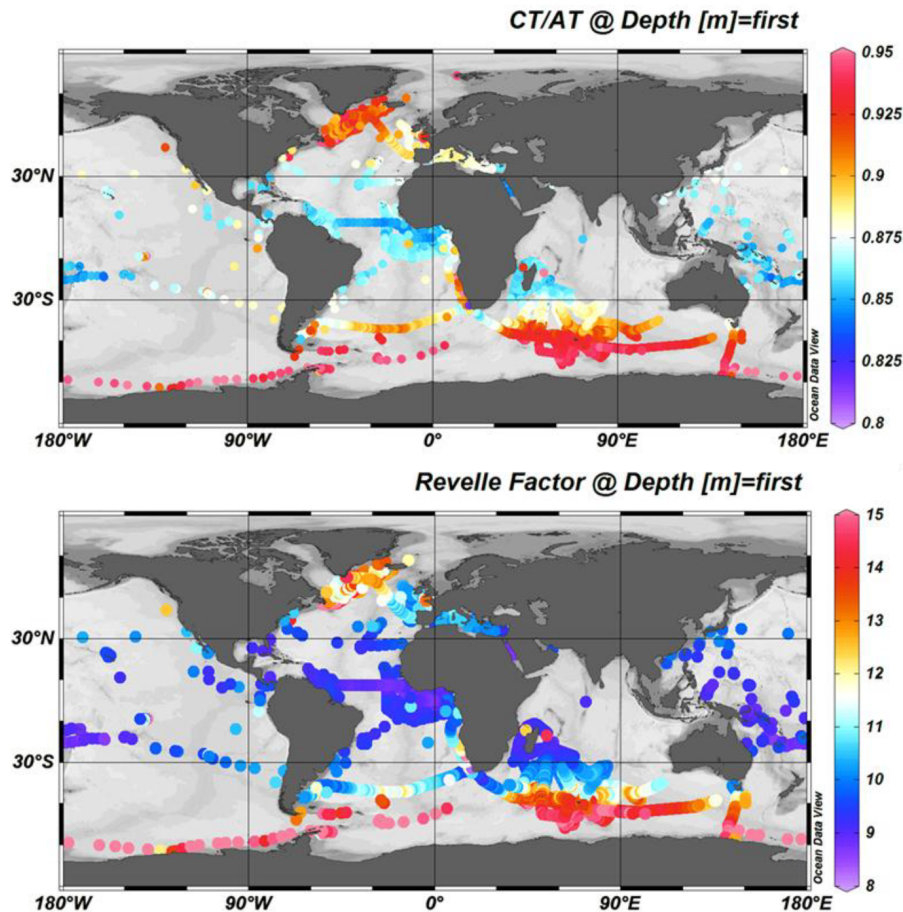
As expected, because of the anthropogenic  $\text{CO}_2$  uptake, the  $C_T$  concentrations increased in most regions (Fig. 12, Table 6). This is identified in the Indian Ocean (in the subtropical and the high-latitude sectors), in the Mediterranean Sea and in coastal waters off Brittany. However, the signal

is more complex in the NASPG. As previously shown, the  $C_T$  trend in the NASPG depends on the season and decade (Metz et al., 2010; Reverdin et al., 2018; Fröb et al., 2019; Leseurre et al., 2020). Here, we selected only the data in January–February from the SURATLANT cruises, leading to a  $C_T$  trend of  $+0.72$  ( $\pm 0.17$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ . Compared with the regions further north, the  $C_T$  trend in the NASPG is about half the  $C_T$  trends of  $+1.44$  ( $\pm 0.23$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  observed in the Iceland Sea (Olafsson et al., 2009) or  $+1.48$  ( $\pm 0.22$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  at station M in the Norwegian Sea (Skjelvan et al., 2022).

In the coastal zones off Brittany, although there are large seasonal and interannual variabilities (Gac et al., 2021), an annual  $C_T$  trend of  $+1.72$  ( $\pm 0.28$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  is detected over 10 years (2009–2019). The same is observed in the Mediterranean Sea, where the  $C_T$  offshore trend of  $+0.69$  ( $\pm 0.18$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  is low compared with what was observed in the coastal zone (SOMLIT-Point-B,  $+2.97$  ( $\pm 0.20$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ ; Kapsenberg et al., 2017).

In the southern Indian Ocean,  $C_T$  concentrations also increased in both the subtropics and at high latitudes,





**Figure 9.** Distribution of the  $C_T/A_T$  ratio (top) and the Revelle factor (bottom) in surface waters (0–10 m). Only data with Flag 2 were used. The figure was produced with ODV (Schlitzer, 2018).

**Table 6.** Trend in  $C_T$  ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ ) and the corresponding standard error in five selected regions where data were available for more than 10 years (data are shown in Fig. 12). The projects/cruises for the selection of the data in each domain are indicated.

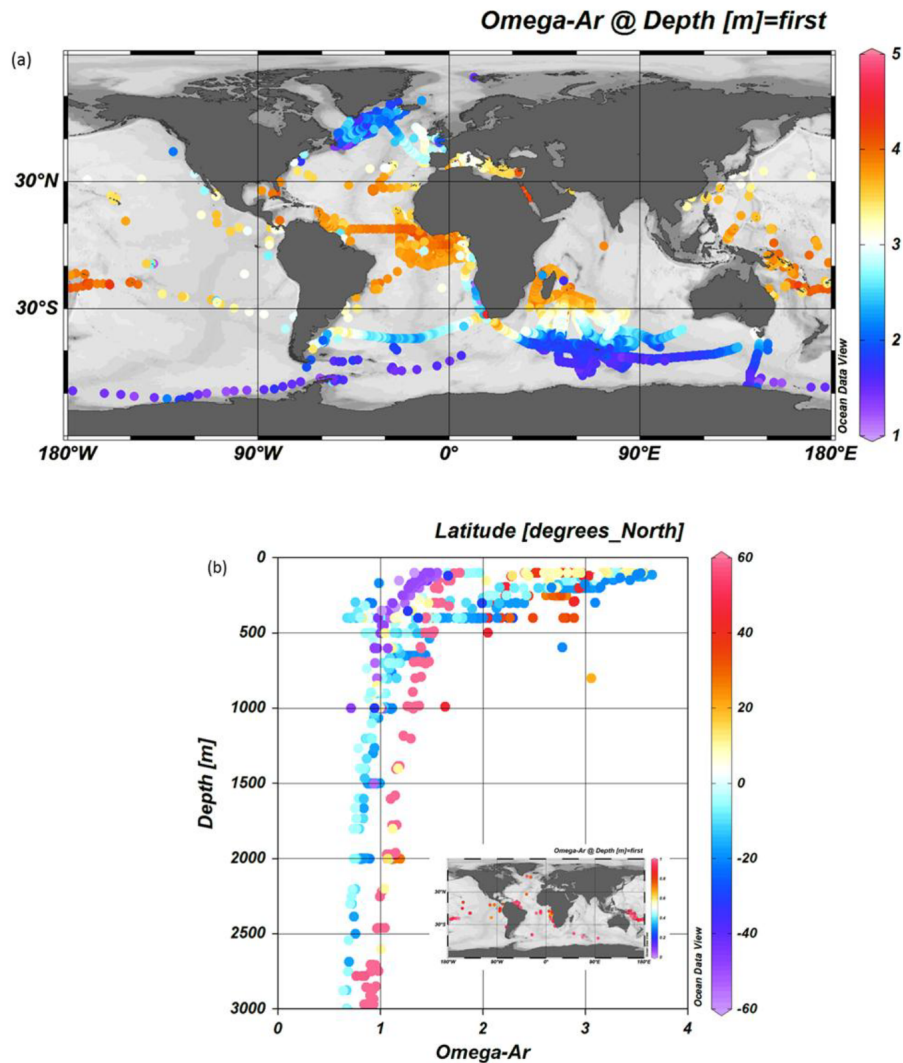
Region (acronym)	Period	$C_T$ trend ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	Season	Projects/Cruises
North Atlantic (NASPG)	1994–2014	+0.72	January–February	SURATLANT
Indian subtropical (IO-SBT)	1998–2018	+0.65	January–February	OISO
Indian south (IO-POOZ)	1998–2018	+0.67	January–February	OISO
Ligurian Sea (MED)	1998–2019	+0.68	All seasons	DYFAMED, BOUSSOLE, MOOSE-GE
Coast of Brittany (BRIT)	2008–2019	+1.72	All seasons	Brest, Roscoff, ECOSCOPA, Penzé

two regions where the primary productivity is relatively low (an oligotrophic regime in the subtropics and a high-nutrient–low-chlorophyll regime, HNLC, south of the polar front). With the data selected for austral summer (January–February), the  $C_T$  trends appeared almost similar in these two regions, around  $+0.65 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  (Table 6).

Finally, in the equatorial Atlantic, the selected data around  $0^\circ$ – $10^\circ$  W highlighted the large variability linked to the oceanic circulation. Detecting a  $C_T$  trend as well as a pos-

sible link to anthropogenic carbon uptake, at least with the data available in 2005–2017, appears to be intricate, as has been previously discussed for the period 2006–2013 (Lefèvre et al., 2016). However, the signal of the  $C_T$  increase is better identified north or south of the Equator in the eastern tropical Atlantic sector (Lefèvre et al., 2021).

In the water column,  $A_T$  and  $C_T$  data from dedicated cruises were used to evaluate the anthropogenic CO<sub>2</sub> ( $C_{\text{ant}}$ ) distribution and pH change since the preindustrial era

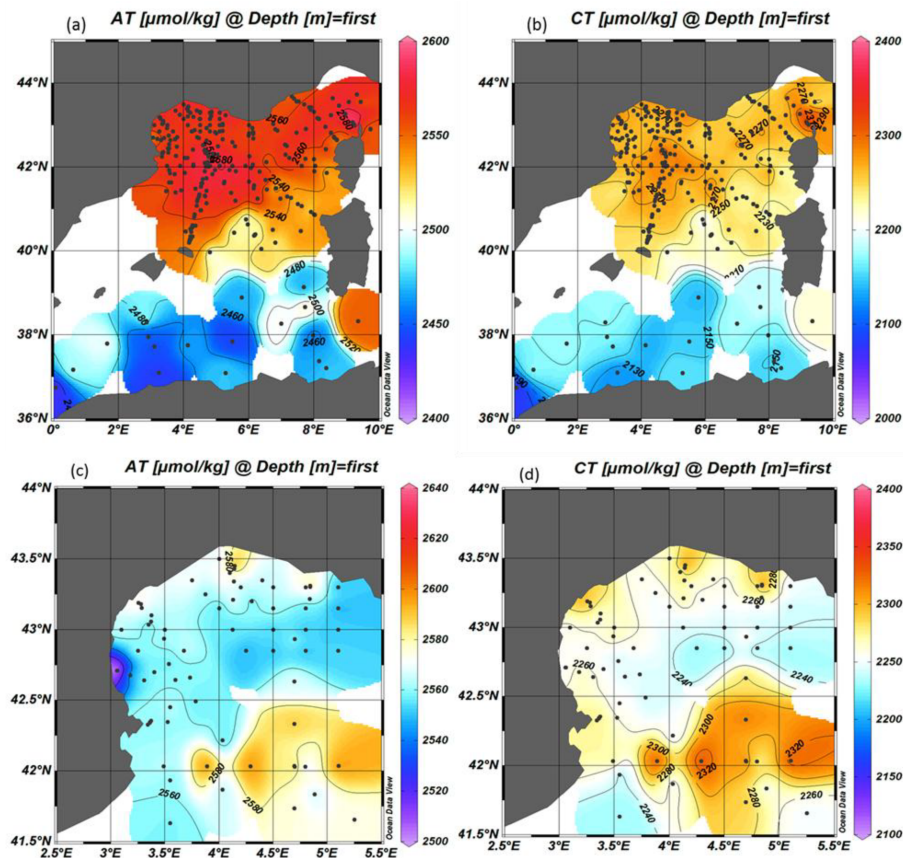


**Figure 10.** Panel (a) shows the distribution of the aragonite saturation state ( $\Omega$ -Ar) in surface waters (0–10 m). Only data with Flag 2 were used. Panel (b) presents depth profiles (100–3000 m) of  $\Omega$ -Ar at few locations in the tropical Pacific, Atlantic and Southern oceans. Stations where undersaturation is detected ( $\Omega$ -Ar < 1) at depth are identified in the inset map. The figure was produced with ODV (Schlitzer, 2018).

(e.g., PANDORA cruise – Ganachaud et al., 2017; OUTPACE cruise – Wagener et al., 2018a; SOMBA cruise – Kerghel et al., 2020). Time series at DYFAMED station also enabled us to investigate the temporal variability in  $C_T$ ,  $A_T$  and  $C_{\text{ant}}$  in the water column (Touratier and Goyet, 2009; Coppola et al., 2020; Fourier et al., 2022). As an example of the observed temporal variations at depth, we selected the data in the 950–1050 m layer in the Ligurian Sea from different cruises (Fig. 13). At that depth, both  $A_T$  and  $C_T$  present some large anomalies that were especially noticeable in 2013 (lower  $A_T$  and  $C_T$  in February 2013, DEWEX cruise) and in 2018 (higher  $A_T$  and  $C_T$  in May 2018, MOOSE-GE cruise), with the latter probably being linked to an episodic convective process that occurred in winter 2018 (Fourrier et al., 2022; Coppola et al., 2023). During the strong convection

event in 2013, the positive anomalies of  $A_T$  and  $C_T$  were mostly identified in the upper layers (Fig. 12c).

In this region, the long-term increase in  $A_T$  indicates that, in addition to the anthropogenic CO<sub>2</sub> signal, other processes are at play to explain the rapid  $C_T$  trend of  $+1.20 (\pm 0.12) \mu\text{mol kg}^{-1} \text{yr}^{-1}$  at depth compared with that observed at the surface (Fig. 12). The signal at depth is probably linked to the variations in the deep convection and mixing with Levantine Intermediate Water (LIW; Margirier et al., 2020) that has higher  $A_T$  and  $C_T$  concentrations. The long-term increase in  $A_T$  and  $C_T$  at depth (here at 1000 m; Fig. 13) was also observed below 2000 m (Coppola et al., 2020); this signal should be investigated using a dedicated analysis with other properties (e.g., O<sub>2</sub> and nutrients), following Fourrier



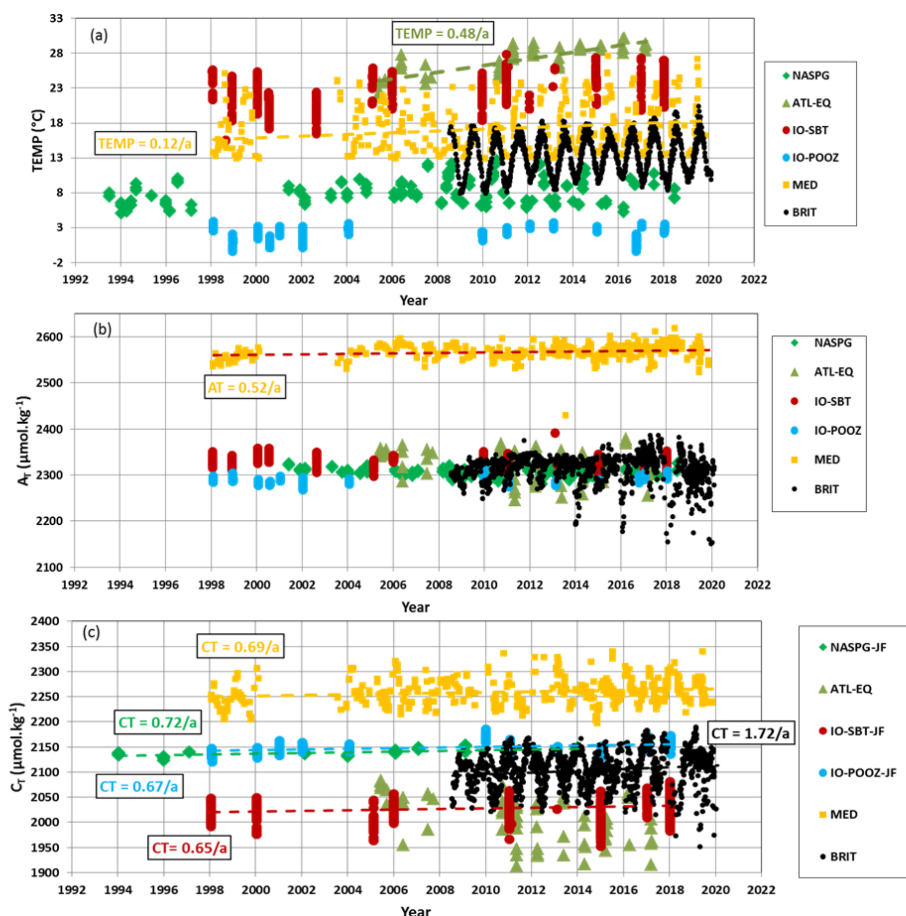
**Figure 11.** Distribution (in  $\mu\text{mol kg}^{-1}$ ) of  $A_T$  (a) and  $C_T$  (b) in surface waters of the western Mediterranean Sea (0–10 m) from all data for 1998–2019. A detailed distribution (in  $\mu\text{mol kg}^{-1}$ ) of  $A_T$  (c) and  $C_T$  (d) in surface waters of the Gulf of Lion is also given for the 2010–2011 period only (the CARBORHONE, CASCADE, LATEX, MOLA and MOOSE-GE cruises). The figure was produced with ODV (Schlitzer, 2018).

et al. (2022) for the 2012–2020 period, and a larger dataset in the Mediterranean Sea (e.g., GLODAP).

## 6 Using $A_T$ and $C_T$ data to validate observations from autonomous instruments

The dataset presented in this synthesis would also offer interesting observations to validate properties ( $A_T$  and  $C_T$ ) derived from Biogeochemical-Argo (BGC-Argo) floats equipped with pH sensors (e.g., Bushinsky et al., 2019; Mazloff et al., 2023; Mignot et al., 2023). The water-column in situ  $A_T$  and  $C_T$  data obtained during the Antarctic Circumpolar Expedition (ACE) in 2016–2017 were collected at a location where Southern Ocean Carbon and Climate Observations and Modeling (SOCCOM) floats were launched (Walton and Thomas, 2018). A SOCCOM float (WMO ID 5905069) was launched on 11 January 2017 at  $55^\circ\text{S}$ ,  $96^\circ\text{E}$ , south of the polar front in the southern Indian Ocean. The pH, temperature and salinity data from the float were then used to derive  $A_T$  and  $C_T$  profiles (here using a multiple linear regression, MLR, algorithm; Williams et al., 2016,

2017). In the top layers, the discrete ACE data (Fig. 14) present large variability in the  $A_T$  and  $C_T$  concentrations that is not captured in the records derived from the float (the MLR method somehow smoothed the profiles). However, given the uncertainty in reconstructed  $A_T$  from float data ( $5.6 \mu\text{mol kg}^{-1}$ ), the average values in the first 100 m were almost identical ( $A_{T-ACE} = 2285.1 (\pm 4.4) \mu\text{mol kg}^{-1}$  and  $A_{T-float} = 2278.3 (\pm 0.7) \mu\text{mol kg}^{-1}$ ;  $C_{T-ACE} = 2139.7 (\pm 9.2) \mu\text{mol kg}^{-1}$  and  $C_{T-float} = 2141.1 (\pm 3.2) \mu\text{mol kg}^{-1}$ ). Moreover, below 200 m, profiles from the float are coherent compared to the  $A_T$  and  $C_T$  measurements (Fig. 14). This is encouraging with respect to using float data to explore the seasonal variability in  $A_T$  and  $C_T$  in the Southern Ocean (e.g., Williams et al., 2018; Johnson et al., 2022) and to estimate anthropogenic  $\text{CO}_2$  in the water column in this sector (Fig. 14). Here, the  $C_{ant}$  concentrations were calculated below 200 m (corresponding to the temperature minimum of the winter in the Southern Ocean and using the TrOCA – Tracer combining Oxygen, inorganic Carbon and total Alkalinity – method; Touratier et al., 2007). The float data suggest that  $C_{ant}$  concentrations are positive down to about 1000 m,



**Figure 12.** Time series of (a) sea-surface temperature ( $^{\circ}\text{C}$ ), (b)  $A_T$  ( $\mu\text{mol.kg}^{-1}$ ) and (c)  $C_T$  ( $\mu\text{mol.kg}^{-1}$ ) in six regions: the North Atlantic subpolar gyre (NASPG, 1993–2018, green diamond), the equatorial Atlantic (ATL-EQ, 2005–2017, green triangle), the Indian subtropical sector (IO-SBT, red circle) and high-latitude sector (IO-POOZ, blue circle) (1998–2018), the Ligurian Sea (MED, 1998–2019, orange square), and times series' stations in the coastal zones off Brittany (BRIT, period 2008–2019, black dots). Trends (dashed lines and values) are shown when relevant for the discussion ( $C_T$  trends listed in Table 6).

with maximum values in the subsurface. In 2017, the mean  $C_{\text{ant}}$  concentration at 200 m was  $49.1 (\pm 9.0) \mu\text{mol.kg}^{-1}$ . Below that depth,  $C_{\text{ant}}$  decreased to  $+29.8 (\pm 8.5) \mu\text{mol.kg}^{-1}$  in the 300–400 m layer. To complement the  $C_{\text{ant}}$  inventories based on the GLODAP data product (e.g., Gruber et al., 2019),  $C_{\text{ant}}$  estimates derived from BGC-Argo floats, as evaluated here in the Southern Ocean, could be applied in other locations, as was previously tested in the North Pacific (Li et al., 2019).

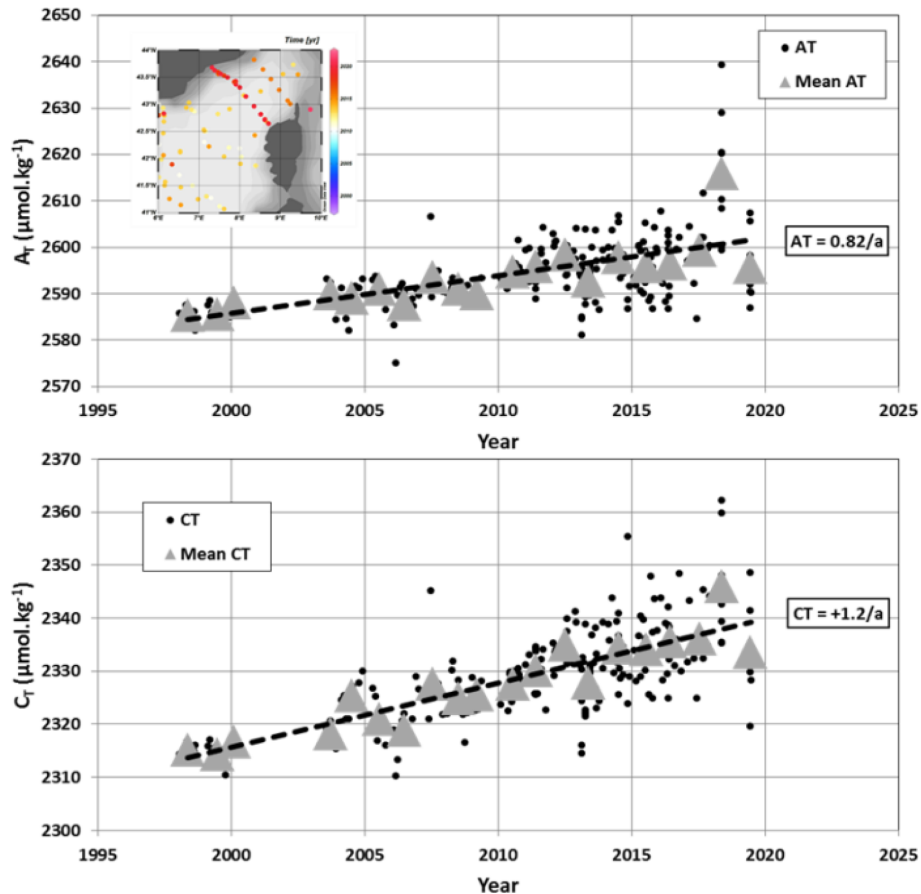
In surface water, as the  $A_T$  derived from the float data are deduced using MLR or LIAR (locally interpolated  $A_T$  regression) methods (Williams et al., 2017; Carter et al., 2016), the  $A_T$  data in the SNAPO-CO<sub>2</sub> synthesis could also be used to identify  $A_T$  anomalies that are not always captured from floats. This is particularly relevant in coccolithophore bloom areas when low  $A_T$  concentrations and high  $p\text{CO}_2$  are observed (e.g., Balch et al., 2016, in the Southern Ocean; Robertson et al., 1994, in the North Atlantic).

## 7 Data availability

Data presented in this study are available from SEANOE: <https://www.seanoe.org> (last access: 22 December 2023), <https://doi.org/10.17882/95414> (Metzl et al., 2023).

## 8 Summary and suggestions

The ocean data synthesized in this product are based on measurements of  $A_T$  and  $C_T$  performed between 1993 and 2022 with an accuracy of  $\pm 4 \mu\text{mol.kg}^{-1}$ . The product offers a large dataset of  $A_T$  and  $C_T$  for the Global Ocean and regional biogeochemical studies. It includes more than 44 400 surface and water-column observations in all oceanic basins, in the Mediterranean Sea, in the coastal zones, near coral reefs and in rivers. For the open ocean, this complements the SOCAT and GLODAP data products (Bakker et al., 2016; Lauvset et al., 2022). For the coastal sites, this also complements the synthesis of coastal time series only done around North



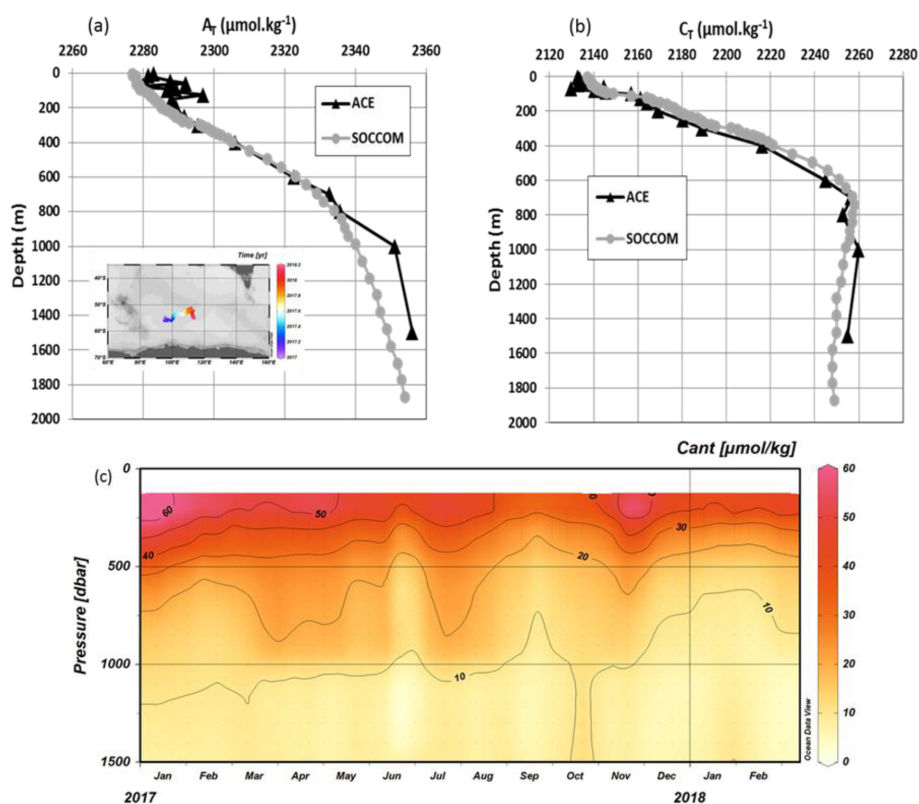
**Figure 13.** Time series of  $A_T$  ( $\mu\text{mol kg}^{-1}$ ) and  $C_T$  ( $\mu\text{mol kg}^{-1}$ ) in the Ligurian Sea (1998–2019) in the 950–1050 m layer. The annual mean (gray triangles) was calculated from all data each year (black dots). The trends (dashed line) based on the annual mean are  $+0.82$  ( $\pm 0.15$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  for  $A_T$  and  $+1.20$  ( $\pm 0.12$ )  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  for  $C_T$ . In this layer, selected data are from the ANTARES, CASCADE, DEWEX, DYFAMED, MOOSE-GE and PEACETIME cruises (location of stations shown in the inset map produced with ODV; Schlitzer, 2018).

America (Fassbender et al., 2018; Jiang et al., 2021; OCADS, 2023).

The SNAPO-CO<sub>2</sub> dataset enables the investigation of seasonal variations to decadal trends in  $A_T$  and  $C_T$  in various oceanic provinces. In regions where data are available for more than 2 decades in surface water (North Atlantic, Ligurian Sea, southern Indian Ocean and coastal regions), all time series show an increase in  $C_T$ . Apart from in the Mediterranean Sea,  $A_T$  appears relatively constant over time, although the  $A_T$  content presents significant interannual variability, such as in the NASPG or in the coastal zones, including near the Congo and Amazon river plumes.

This dataset represents independent data for the validation of reconstructed  $A_T$  or  $C_T$  fields using various methods (e.g., Rödenbeck et al., 2013, 2015; Sauzède et al., 2017; Turk et al., 2017; Bittig et al., 2018; Broullón et al., 2019, 2020; Land et al., 2019; Keppler et al., 2020; Fourier et al., 2020; Gregor and Gruber, 2021; Sims et al., 2023). It is also useful to validate Earth system models (ESMs) that currently

present bias with respect to the reproduction of the seasonal cycle of  $C_T$  and  $A_T$  due to the inadequate representation of biogeochemical cycles, including the coupling of biological and physical processes (e.g., Pilcher et al., 2015; Mongwe et al., 2018; Lerner et al., 2021). This should be resolved in order to ensure confident future projections of productivity, ocean acidification and the responses of the marine ecosystems (e.g., Kwiatowski et al., 2020). Recall that ocean biogeochemical global models (OBGMs) or ESMs calculate  $p\text{CO}_2$  from  $A_T$ – $C_T$  pairs and that the simulated annual  $\text{CO}_2$  flux might be correct when compared to observations but for the wrong reasons (e.g., Goris et al., 2018; Lerner et al., 2021). For example, it has been shown that biases in  $A_T$  in ESMs lead to an overestimation of the oceanic  $f\text{CO}_2$  trend and, thus, uncertainty when predicting the oceanic anthropogenic  $\text{CO}_2$  uptake (Lebehot et al., 2019). The simulated seasonal cycle of  $f\text{CO}_2$  is also uncertain in ESMs, especially at high latitudes (e.g., Joos et al., 2023). Thus, it is important



**Figure 14.** Profiles of (a)  $A_T$  ( $\mu\text{mol.kg}^{-1}$ ) and (b)  $C_T$  ( $\mu\text{mol.kg}^{-1}$ ) observed at station ACE-20 (55° S, 95° E; 11 January 2017, black triangles) compared with the profiles deduced from the SOCCOM float (WMO code 5905069) launched at that location (first data on 12 January 2017, gray circles). The location and drift of the float in 2017–2018 are shown on the inset map. Panel (c) presents a Hovmöller section (pressure versus time) of anthropogenic CO<sub>2</sub> concentrations ( $C_{\text{ant}}$ , in  $\mu\text{mol.kg}^{-1}$ ) estimated from the float data ( $A_T$ ,  $C_T$ ,  $O_2$ ,  $T$ ) below 200 m (period from January 2017 to February 2018). The section was produced with ODV (Schlitzer, 2018).

to attempt to validate ESMs with  $A_T$  and  $C_T$  data, such as undertaken in this synthesis.

This dataset would also serve to validate autonomous platforms capable of measuring pH and  $f\text{CO}_2$  variables and, along with SOCAT and GLODAP datasets, provides an additional reference dataset for the development and validation of regional biogeochemical models for simulating air–sea CO<sub>2</sub> fluxes. It is also essential for training and validating neural networks capable of predicting variables in the carbonate system, thereby enhancing observations of marine CO<sub>2</sub> at different spatial and temporal scales.

The data presented here are available online on the SEANOE server (Metzl et al., 2023, <https://doi.org/10.17882/95414>) and are divided in two files: one for the Global Ocean and one for the Mediterranean Sea. The sources of the original datasets (DOI) and their associated references are listed in the Supplement (Tables S3, S4). We invite users to comment on any anomalies that have not been detected or to suggest any potential misqualifications of data in the present product (e.g., data probably good, although assigned Flag 3, probably wrong). The SNAPO-CO<sub>2</sub> dataset will be regularly updated on the

SEANOE data server (with new observations), controlled and archived.

**Supplement.** The supplement related to this article is available online at: <https://doi.org/10.5194/essd-16-89-2024-supplement>.

**Author contributions.** NM prepared the data synthesis, the figures and wrote the draft of the manuscript with contributions from all authors. JF measured the discrete samples (from 2014 on) with help from CM and CLM and also prepared the individual reports for each project. NM and JF pre-qualified the discrete  $A_T$  and  $C_T$  data. CLM and NM are co-investigators of the ongoing OISO project and qualified the underway  $A_T$  and  $C_T$  data from OISO and CLIM-EPARSES cruises. All authors have reviewed the manuscript and contributed to one or more of the following: organizing cruises, sample collection and/or data qualification.

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