

1 An updated synthesis of ocean total alkalinity and dissolved inorganic carbon measurements  
2 from 1993 to 2023: the SNAPO-CO2-v2 dataset

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36

37 **Abstract.** Total alkalinity ( $A_T$ ) and dissolved inorganic carbon ( $C_T$ ) in the oceans are important properties to  
38 understand the ocean carbon cycle and its link with global change (ocean carbon sinks and sources, ocean  
39 acidification) and ultimately find carbon based solutions or mitigation procedures (marine carbon removal). We  
40 present an extended database (SNAPO-CO2, Metzl et al, 2024d) with 24700 new additional data for the period  
41 2002 to 2023. The full database now includes more than 67000  $A_T$  and  $C_T$  observations along with basic  
42 ancillary data (time and space location, depth, temperature and salinity) in various oceanic regions obtained since  
43 1993 mainly in the frame of French research projects. This includes both surface and water columns data  
44 acquired in open oceans, coastal zones, rivers and in the Mediterranean Sea and either from time-series or  
45 punctual cruises. Most  $A_T$  and  $C_T$  data in this synthesis were measured from discrete samples using the same  
46 closed-cell potentiometric titration calibrated with Certified Reference Material, with an overall accuracy of  $\pm 4$   
47  $\mu\text{mol kg}^{-1}$  for both  $A_T$  and  $C_T$ . The same technique was used onboard for underway measurements during cruises  
48 conducted in the Southern Indian and Southern Oceans. The  $A_T$  and  $C_T$  data from these cruises are also added in

49 this synthesis. The data are provided in one dataset for the global ocean (<https://doi.org/10.17882/102337>) that  
50 offers a direct use for regional or global purposes, e.g.  $A_T$ /Salinity relationships, long-term  $C_T$  estimates,  
51 constraint and validation of diagnostics  $C_T$  and  $A_T$  reconstructed fields or ocean carbon and coupled  
52 climate/carbon models simulations, as well as data derived from Biogeochemical-Argo (BGC-Argo) floats.  
53 These data can also be used to calculate pH, fugacity of  $\text{CO}_2$  ( $f\text{CO}_2$ ) and other carbon system properties to derive  
54 ocean acidification rates or air-sea  $\text{CO}_2$  fluxes.

55

## 56 **1 Introduction**

57

58 The ocean plays a major role in reducing the impact of climate change by absorbing more than 90% of  
59 the excess heat in the climate system (Cheng et al., 2020, 2024; von Schuckmann et al, 2023; IPCC, 2022) and  
60 about 25% of human released  $\text{CO}_2$  (Friedlingstein et al., 2022, 2023). In the last decade, the oceans experienced  
61 a rapid warming, the year 2023 being the hottest since 1955 (Cheng et al, 2024). In the atmosphere the  $\text{CO}_2$   
62 concentration continues its terrific progressive rising, reaching 419.3 ppm in 2023 (a rate of +2.83 ppm yr<sup>-1</sup>, Lan  
63 et al 2024). In August 2024, the global atmospheric  $\text{CO}_2$  concentration was already above 420 ppm. In the next  
64 decade the oceans will continue to capture heat and  $\text{CO}_2$ , somehow limiting the climate change, but this oceanic  
65  $\text{CO}_2$  uptake changes the chemistry of seawater reducing its buffering capacity (Revelle and Suess, 1957; Jiang et  
66 al, 2023). This process known as ocean acidification has potential impacts on marine organisms (Fabry et al.,  
67 2008; Doney et al., 2009, 2020; Gattuso et al., 2015). With atmospheric  $\text{CO}_2$  concentrations, surface ocean  
68 temperature and ocean heat content, sea-level, sea-ice and glaciers, the ocean acidification (decrease of pH) is  
69 now recognized by the World Meteorological Organization as one of the 7 key properties for global climate  
70 indicators (WMO, 2018). Ocean acidification is specifically referred in the SDG indicator 14.3.1 coordinated at  
71 the Intergovernmental Oceanographic Commission (IOC) of UNESCO. Observing the carbonate system in the  
72 open oceans, coastal zones and marginal seas and understanding how this system changes over time is thus  
73 highly relevant not only to quantify the global ocean carbon budget, the anthropogenic  $\text{CO}_2$  inventories or ocean  
74 acidification rates, but also to understand and simulate the processes that govern the complex  $\text{CO}_2$  cycle in the  
75 ocean (e.g. Goyet et al, 2016, 2019) and to better predict the future evolution of climate and global changes  
76 (Eyring et al., 2016; Kwiatkowski et al., 2020; Jiang et al., 2023). As the rate of change in ocean acidification  
77 presents large temporal and regional variability, long-term observations are required. Weekly to monthly regular  
78 resolution data are needed to better investigate the long-term change of the carbonate system in regions subject  
79 to extreme events (e.g. tropical cyclones, marine heat or cold waves, rapid freshening, convection, dust events,  
80 river discharges, etc....). In this context it is recommended to progress in data synthesis of the ocean carbon  
81 observations that would offer new high quality products for the community (e.g. for GOA-ON, [www.goa-on.org](http://www.goa-on.org),  
82 IOC/SDG 14.1.3, <https://oa.iode.org/>, Tilbrook et al., 2019).

83 In this work, following the first SNAPO- $\text{CO}_2$  synthesis product (Metzl et al, 2024a), we present a new  
84 synthesis of more than 67000  $A_T$  and  $C_T$  data, measured either on shore or onboard Research Vessels obtained  
85 over the 1993-2023 period during various cruises or at time-series stations mainly supported by French projects.  
86 Hereafter this new dataset will be cited as SNAPO- $\text{CO}_2$ -v2. The methods, data assemblage and quality control  
87 were presented in version V1. Here, we describe the new data added and discuss some potential uses of this  
88 dataset.

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## 90 2 Data collections

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92 The time series projects and research cruises from which new data were collated are listed in Table 1  
93 with information and references in the Supplementary file (Tables S1, S3 and S4). The sampling locations of  
94 new data are displayed in Figure 1 (the location for all data presented in Figure S1). Sampling was performed  
95 either from CTD-Rosette casts (Niskin bottles) or from the ship's seawater supply (intake at about 5m depth  
96 depending on the ship and swell). Samples collected in 500 mL borosilicate glass bottles were poisoned with 100  
97 to 300  $\mu\text{L}$  of  $\text{HgCl}_2$  depending on the cruises, closed with greased stoppers (Apiezon®) and held tight using  
98 elastic band following the SOP protocol (DOE, 1994; Dickson et al., 2007). Some samples were also collected in  
99 500 mL bottles closed with screw caps. After completion of each cruise, most of discrete samples were returned  
100 back to the LOCEAN laboratory (Paris, France) and stored in a dark room at 4 °C before analysis generally  
101 within 2-3 months after sampling (sometimes within a week). In this version we added data from samples that  
102 were also returned to University of Perpignan or to University of La Réunion. In addition to discrete samples  
103 analyzed for various projects conducted mainly in the North Atlantic, Tropical Atlantic, Mediterranean Sea and  
104 coastal regions (Table 1), we complemented this second synthesis with  $A_T$  and  $C_T$  surface observations obtained  
105 in the Indian and Southern oceans during the OISO cruises in 2019-2021 (Leseurre et al., 2022; Metzl et al,  
106 2022; data also available at NCEI/OCADS: [www.nodc.noaa.gov/ocads/oceans/VOS\\_Program/OISO.html](http://www.nodc.noaa.gov/ocads/oceans/VOS_Program/OISO.html)) and  
107 MINERVE cruises in 2002-2018 (Laika et al, 2009; Brandon et al, 2022). The  $A_T$  and  $C_T$  measurements from the  
108 MINERVE cruises were performed either onboard R/V Astrolabe or back in the laboratories (at LOCEAN  
109 laboratory and at University of Perpignan).

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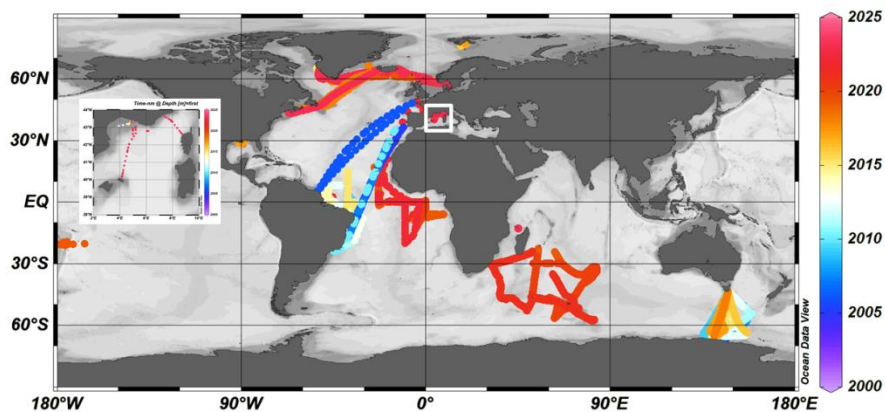
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**Figure 1:** Locations of new  $A_T$  and  $C_T$  data (2005-2023) in the Global Ocean and the Western Mediterranean Sea (white box, insert) in the SNAPO-CO2-v2 dataset. Color code is for Year. Figure produced with ODV (Schlitzer, 2018).

125 **Table 1:** List of cruises added in the SNAPO-CO2-v2 dataset. This is organized by region from North to South  
 126 and the Mediterranean Sea. See Tables S1, S2, S3 and S4 in the Supplementary Material for a list of laboratories,  
 127 of CRMs used, for DOI and for references of cruises. Nb = the number of data for each cruise or time-series. \*  
 128 indicates the measurements at sea (surface underway).

Cruise/Project	Start	End	Region	Sampling	Nb	
134	STEP	2016	2017	Arctic	Water Column	33
135	SURATLANT AX1	2017	2023	North Atlantic	Surface	255
136	SURATLANT AX2	2018	2023	North Atlantic	Surface	224
137	VOS	2005	2010	Atlantic	Surface	192
138	MISSRHODIA-1	2017	2017	Gulf Mexico	Water Column	8
139	ACIDHYPO	2022	2022	Gulf Mexico	Water Column	10
140	CAMFIN-WATL	2010	2015	Trop Atlantic	Surface	192
141	PIRATA-BR	2009	2015	Trop Atlantic	Surface	194
142	BIOAMAZON	2013	2014	Trop Atlantic	Surface	62
143	AMAZOMIX	2021	2021	Trop Atlantic	Water Column	180
144	PIRATA-FR	2019	2019	Trop Atlantic	Surface	93
145	PIRATA-FR	2020	2020	Trop Atlantic	Surface, Water Column	58
146	PIRATA-FR	2021	2021	Trop Atlantic	Surface, Water Column	79
147	PIRATA-FR	2022	2022	Trop Atlantic	Surface, Water Column	118
148	CO2ARVOR	2009	2010	Atlantic, Coastal	Surface, Water Column	621
149	SOMLIT-Roscoff	2020	2022	Coastal North Atl	Surface and 60m	207
150	SOMLIT-Brest	2020	2022	Coastal North Atl	Surface	251
151	TONGA	2019	2019	Trop Pacific	Water Column	226
152	CARBODISS	2018	2019	Indian Mayotte	Surface	85
153	OISO *	2019	2021	South Indian	Surface	5258
154	MINERVE	2004	2018	Southern Ocean	Surface	1077
155	MINERVE *	2002	2013	Southern Ocean	Surface	11258
156	COCORICO2	2017	2022	Coastal	Surface	589
157	SOMLIT-PointB	2019	2023	MedSea Coastal	Surface and 50m	716
158	SOLEMIO	2018	2022	MedSea Coastal	Water Column	271
159	ANTARES	2017	2023	MedSea	Water Column	506
160	MOLA	2018	2023	MedSea Coastal	Water Column	193
161	DYFAMED	2018	2023	MedSea	Water Column	514
162	MESURHO-BENT	2010	2011	MedSea Coastal	Surface and sub-surface	25
163	ACCESS-01	2012	2012	MedSea Coastal	Water Column	16
164	CARBO-DELTA-2	2013	2013	MedSea Coastal	Water Column	14
165	DICASE	2014	2014	MedSea Coastal	Water Column	22
166	MISSRHODIA-2	2018	2018	MedSea Coastal	Surface and sub-surface	13
167	DELTARHONE1	2022	2022	MedSea Coastal	Water Column	9
168	MOOSE-GE	2021	2021	MedSea	Water Column	451
169	MOOSE-GE	2022	2022	MedSea	Water Column	447
170	MOOSE-GE	2023	2023	MedSea	Water Column	475

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### 173 3 Method, accuracy, repeatability and quality control

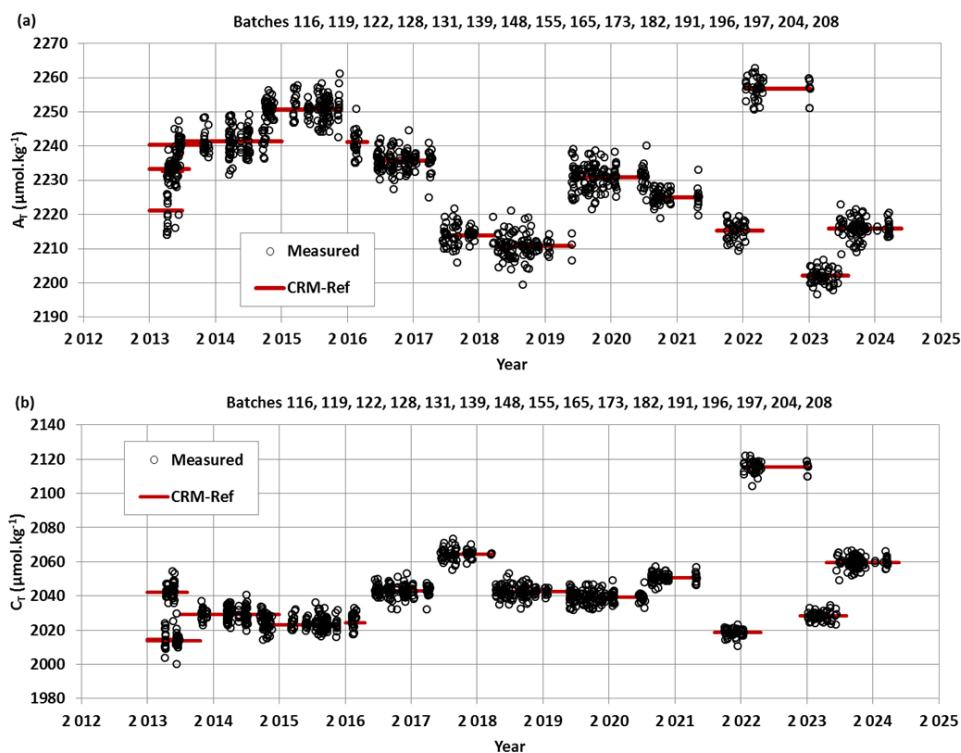
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#### 175 3.1 Method and accuracy

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177 Since 2003, the discrete samples returned back at SNAPO-CO2 Service facilities (LOCEAN, Paris),  
 178 were analyzed simultaneously for  $A_T$  and  $C_T$  by potentiometric titration using a closed cell (Edmond, 1970;  
 179 Goyet et al., 1991). The same technique was used at sea for surface water underway measurements during OISO  
 180 and MINERVE cruises (indicated by \* in Table 1). In the late 1980s the so-called “JGOFS-IOC Advisory Panel  
 181 on Ocean CO2” recommended the need for standard analysis protocols and for developing Certified Reference  
 182 Materials (CRMs) for inorganic carbon measurements (Poisson et al., 1990; UNESCO, 1990, 1991). The CRMs

183 were provided to international laboratories by Pr. A. Dickson (Scripps Institution of Oceanography, San Diego,  
 184 USA), starting in 1990 for  $C_T$  and 1996 for  $A_T$ , respectively. These CRMs were thus always available to us and  
 185 used to calibrate the measurements (CRM Batch numbers used for each cruise are listed in the Supplementary  
 186 file, Table S2). The CRMs accuracy, as indicated in the certificate for each Batch, is around  $\pm 0.5 \mu\text{mol kg}^{-1}$  for  
 187 both  $A_T$  and  $C_T$  ([www.nodc.noaa.gov/ocads/oceans/Dickson\\_CRM/batches.html](http://www.nodc.noaa.gov/ocads/oceans/Dickson_CRM/batches.html)). The concentrations of CRMs  
 188 we used vary between 2193 and 2426  $\mu\text{mol kg}^{-1}$  for  $A_T$  and between 1968 and 2115  $\mu\text{mol kg}^{-1}$  for  $C_T$   
 189 corresponding to the range of concentrations observed in open ocean water. In the Mediterranean Sea the  
 190 concentrations are higher ( $A_T > 2600 \mu\text{mol kg}^{-1}$  and  $C_T > 2300 \mu\text{mol kg}^{-1}$ ) and in the coastal zones or near the  
 191 Amazon River plume the concentrations were often lower than the CRMs ( $A_T < 1500 \mu\text{mol kg}^{-1}$  and  $C_T < 1000$   
 192  $\mu\text{mol kg}^{-1}$ ). Results of analyses performed on 1242 CRM bottles (different Batches) in 2013-2024 are presented  
 193 in Figure 2. The standard-deviations (Std) of the differences of measurements were on average  $\pm 2.69 \mu\text{mol kg}^{-1}$   
 194 for  $A_T$  and  $\pm 2.88 \mu\text{mol kg}^{-1}$  for  $C_T$ . For unknown reasons, the differences were occasionally up to 10-15  $\mu\text{mol kg}^{-1}$   
 195  $^{-1}$  (1.2% of the data, Figure S2). These few CRM measurements were discarded for the data processing. We did  
 196 not detect any specific signal for CRM analyses (e.g., larger uncertainty depending on the Batch number or  
 197 temporal drifts during analyses, Figure 2) but for some cruises the accuracy based on CRMs could be better than  
 198 3  $\mu\text{mol kg}^{-1}$  (e.g.  $< 3 \mu\text{mol kg}^{-1}$  for AMAZOMIX cruise using 6 Batches #197 and for MOOSE-GE 2022 using  
 199 19 Batches #204, or  $< 1.5 \mu\text{mol kg}^{-1}$  for SOMLIT-Point-B in 2022 using 6 Batches #204).



220 **Figure 2:**  $A_T$  (a) and  $C_T$  (b) analyses for different CRM Batches measured in 2013-2024. For these 1242  
 221 analyses the mean and standard-deviations of the differences with the CRM reference were  $-0.11 (\pm 2.69) \mu\text{mol}$   
 222  $\text{kg}^{-1}$  for  $A_T$  and  $0.01 (\pm 2.88) \mu\text{mol kg}^{-1}$  for  $C_T$ .

### 226 3.2 Repeatability

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228 For some projects, duplicates have been regularly sampled (SOMLIT-Point-B, SOMLIT-Brest) or  
229 replicate bottles sampled at selected depths at fixed stations during the cruises (e.g. STEP, CARBODISS). In the  
230 first synthesis of the SNAPO-CO<sub>2</sub> dataset we showed the results from several time-series (SOMLIT-Point-B,  
231 SOMLIT-Brest and BOUSSOLE/DYFAMED). Here we present the results for the new data obtained at  
232 SOMLIT-Point-B in the coastal Mediterranean Sea and SOMLIT-Brest in the Bay of Brest (Figure 3). Results of  
233  $A_T$  and  $C_T$  repeatability are synthesized in Table 2. For the OISO cruises conducted in 2019, 2020 and 2021 the  
234 repeatability was evaluated from duplicate analyses (within 20 minutes time) of continuous sea surface  
235 underway sampling at the same location (when the ship was stopped). Similarly to what was found for the CRM  
236 measurements (Figure S2), differences in duplicates are occasionally higher than 10-15  $\mu\text{mol kg}^{-1}$  (Figure 3) but  
237 most of the duplicates for all projects are within 0 to 3  $\mu\text{mol kg}^{-1}$ . Compared to previous results (Kapsenberg et  
238 al. 2017; Metzl et al, 2024a), there are larger differences between duplicates at SOMLIT-B in 2019-2023 (up to  
239 30  $\mu\text{mol kg}^{-1}$ , Figure 3) leading to relatively large Std around 5 and 6  $\mu\text{mol kg}^{-1}$  for both  $A_T$  and  $C_T$  (Table 2).  
240 The same was observed for duplicates at SOMLIT-Brest (Table 2). We do have not yet a clear explanation for  
241 this large Std although larger variability was observed in recent years, and the measurements were performed  
242 later after the sampling (e.g. more than 6 months for some samples during and after the COVID period). We will  
243 see that given the temporal variability of the properties this does not lead to suspicious interpretation for the  
244 seasonality or the trend analyses of these time-series.

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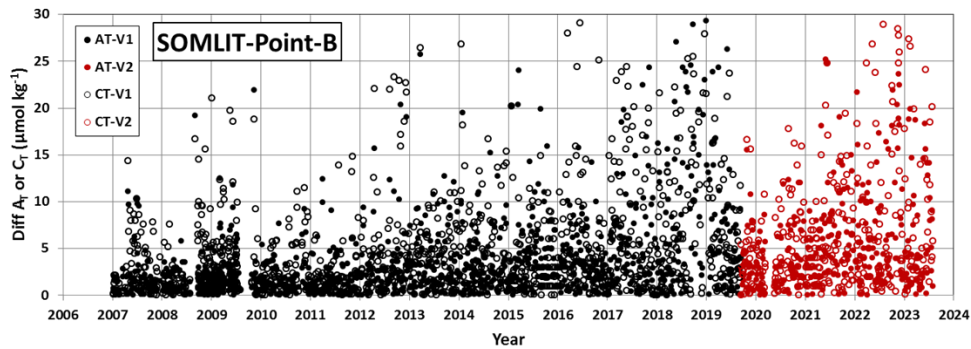
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**Figure 3:** Results of duplicate  $A_T$  and  $C_T$  analyses from the time-series SOMLIT-Point-B in the coastal Mediterranean Sea and SOMLIT-Brest in the coastal Brittany for the data in the SNAPO-CO<sub>2</sub>-v1 dataset (black) and new data added in SNAPO-CO<sub>2</sub>-v2 (red). The plots show differences in duplicates for both  $A_T$  (filled circles) and  $C_T$  (open circles). Standard-deviations of these duplicates are listed in Table 2.

269 **Table 2:** Repeatability of  $A_T$  and  $C_T$  analyses for cruises with duplicate analysis. The results are expressed as the  
 270 standard-deviations (Std) of the analysis of replicated samples. Nb = the number of replicates for each Time-  
 271 series or Cruise. For the OISO cruises the mean repeatability was obtained from measurements at the same  
 272 location (when the ship stopped).

Cruise	Period	Nb	Std $A_T$ $\mu\text{mol kg}^{-1}$	Std $C_T$ $\mu\text{mol kg}^{-1}$	Reference
STEP	2017	3	0.7	2.8	Unpublished
CARBODISS	2018	10	6.72	5.71	Unpublished
SOMLIT-Point-B	2007-2019	1130	4.5	5.1	SNAPO-CO2-v1, (a)
SOMLIT-Point-B	2019-2023	321	5.2	6.2	SNAPO-CO2-v2, (a)
SOMLIT-Brest	2008-2018	404	3.1	3.4	SNAPO-CO2-v1, (a)
SOMLIT-Brest	2019-2022	142	6.0	6.1	SNAPO-CO2-v2, (a)
OISO 29	2019	46	1.8	1.8	Leseurre et al (2022), (b)
OISO 30	2020	67	1.5	2.0	Metzl et al. (2022), (b)
OISO 31	2021	343	2.6	3.3	Metzl et al (2024c), (b)

292 (a) See Figure 3 for the results of regular duplicates for time-series SOMLIT-Point-B, SOMLIT-Brest.

293 (b) Metadata and data available at [www.nodc.noaa.gov/ocads/oceans/VOS\\_Program/OISO.html](http://www.nodc.noaa.gov/ocads/oceans/VOS_Program/OISO.html)

### 295 3.3 Assigned flags for quality control

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 297 Identifying each data with an appropriate flag is very convenient for selecting the data (good,  
 298 questionable or bad). Here we used 4 flags for each property (flags 2 = good, 3= questionable, 4=bad, and 9= no  
 299 data) following the WOCE program and used in other data products such as SOCAT (Bakker et al., 2016) or  
 300 GLODAP (Olsen et al., 2016; Lauvset et al., 2024). During the data-processing, we first assigned a flag for each  
 301  $A_T$  and  $C_T$  data based on the standard error in the calculation of  $A_T$  and  $C_T$  concentrations (non-linear regression,  
 302 Dickson et al. 2007). By default, if the standard deviation on the regression is  $> 1 \mu\text{mol kg}^{-1}$ , we assigned a flag 3  
 303 (questionable) although the data could be acceptable and then used for interpretations. Flag 3 was also assigned  
 304 when salinity was doubtful or when differences of duplicates were large (e.g.  $\pm 20 \mu\text{mol kg}^{-1}$ ). Flags 4 (bad or  
 305 certainly bad) were assigned when clear anomalies were detected for unknown reasons (e.g. a sample probably  
 306 not fixed with  $\text{HgCl}_2$  or analysis performed late during the COVID issue). A secondary quality control was  
 307 performed by the PIs of each project based on data inspection, duplicates,  $A_T$ /Salinity relationship, or the mean  
 308 observations in deep layers where large variability in  $A_T$  and  $C_T$  is unlikely to occur from year to year.

309 An example for quality flag is presented for all data from the MINERVE cruises conducted in 2002-  
 310 2018 in the Southern Ocean where clear outliers have been identified (Figure S3). For the MINERVE cruises in  
 311 2002-2018 and a total of 12335  $A_T$  and  $C_T$  analyses, 24 were identified as bad (flag 4), 978 for  $A_T$  and 971 for  $C_T$   
 312 listed as questionable (flag 3), and all others are considered as good data (flag 2, i.e. about 92%). For the  
 313 MOOSE-GE cruises in 2021, 2022 and 2023 (new data in SNAPO-CO2-v2) and a total of 1373  $A_T$  and  $C_T$   
 314 analyses, 2 were identified flagged as bad (flag 4), 38 for  $A_T$  and 33 for  $C_T$  listed as questionable (flag 3) all  
 315 others were considered as good data (flag 2, i.e. 97%). This is better than the statistics we evaluated for the  
 316 SNAPO-CO2-v1 dataset (90% flag 2 for MOOSE-GE in 2010-2019). A similar control was performed for each  
 317 project.

### 318 **3.4 Inter-comparisons**

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320 Inter-comparisons of measurements performed for different cruises or with different techniques help to  
321 evaluate the quality of the data and detect potential biases when merging the data in the same region obtained by  
322 different laboratories at different periods. This is especially important to interpret long-term trends of  $A_T$  and  $C_T$   
323 as well as for  $pCO_2$  and pH calculated with  $A_T$   $C_T$  pairs. The synthesis of various cruises in the same region and  
324 periods also offers verification and secondary control of the data.

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#### 326 **3.4.1 Comparisons in deep layers**

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328 Comparisons of data in the deep layers from different cruises are useful for secondary quality control as  
329 one expects low natural variability or anthropogenic signals from season to season and over a few years. Several  
330 cruises were conducted in the Mediterranean Sea in 2017-2023 (MOOSE-GE, ANTARES and DYFAMED). The  
331 mean values of  $C_T$  and  $A_T$  in the deep layers (> 1800m) for each cruise confirmed the coherence of the data  
332 (Table 4). The  $C_T$  and  $A_T$  concentrations are also in the range of the mean values evaluated for cruises conducted  
333 in 2014 in the Mediterranean Sea (results listed in the SNAPO-CO2-v1 synthesis, Metzl et al, 2024a). In the  
334 western tropical Pacific we also observed coherent properties for the TONGA and OUTPACE cruises (Wagener  
335 et al, 2018) for data selected at 1800-2300m layer corresponding to the  $C_T$  maximum layer in the Pacific Deep  
336 Water (PDW). On the other hand in the western tropical Atlantic near the Amazon River plume where the spatial  
337 variability of the properties is large at the surface (Ternon et al, 2000; Mu et al, 2021; Olivier et al, 2022) the  
338 comparison in the water column is less clear (Figure S4). Nevertheless for the AMAZOMIX and the TARA-  
339 Microbiome cruises, both conducted in September 2021, the results at close stations (around 5°N/50°W) suggest  
340 very similar concentrations at 1000m (Table 4). The comparisons in deep waters enabled to merge the different  
341 datasets for interpretations of the temporal trends and processes driving the  $CO_2$  cycle in these regions (e. g.  
342 Ulses et al., 2023 and Wimart-Rousseau et al., 2023 for the Mediterranean Sea)

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#### 344 **3.4.2 Comparing on board and on shore results**

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346 In surface waters where the variability is high inter-comparison is not relevant for secondary quality  
347 control. However, during the MINERVE cruises, discrete samples were occasionally performed along with sea  
348 surface underway measurements. Thus, we can compare  $A_T$  and  $C_T$  measured in the laboratory with those  
349 measured onboard as described by Laika et al. (2009) for the MINERVE cruises in 2005-2006. It should be  
350 noticed that the discrete samples were measured after a long trip (shipping boxes from Hobart, Tasmania to  
351 Paris, France) and thus generally analyzed at least 3 months after the cruises (cruises conducted in October to  
352 February, analyses performed in May-June). Given all the uncertainties associated to the sampling, samples  
353 storage and transport, analyses and CRMs, the mean differences between discrete and underway data are still  
354 reasonable (Std ranging between 4 and 12  $\mu\text{mol kg}^{-1}$ , Table 5). For unknown reasons the mean difference was  
355 high for a cruise in 2008-2009 (Std > 10  $\mu\text{mol kg}^{-1}$ , the “weather goal”, Newton et al., 2015). With this in mind,  
356 we believe the MINERVE data (both underway and discrete data) are useful to interpret the change of properties  
357 in this region at seasonal or decadal scales (Laika et al., 2009; Brandon et al., 2022).

358



359 **Table 4:** Mean observations in the deep layers (> 1800m) of the Ligurian Sea (Western Mediterranean Sea for  
360 different cruises conducted in 2017-2023), of the Tropical Pacific (around 2000m for cruises in 2017 and 2019),  
361 and of the Tropical Atlantic (around 1000m for cruises in 2021).  $N-A_T$  and  $N-C_T$  are  $A_T$  and  $C_T$  normalized at  
362 salinity ( $S = 38$  in the Ligurian Sea;  $S = 35$  for the Pacific and the Atlantic Oceans). Nb = number of data (with  
363 flag 2). Standard deviations are in brackets.

Cruise	Period	Nb	Pot. Temp (°C)	Salinity	$N-A_T$ ( $\mu\text{mol kg}^{-1}$ )	$N-C_T$ ( $\mu\text{mol kg}^{-1}$ )
Ligurian Sea (> 1800m)						
All Cruises	2017-2023	227	12.923 (0.052)	38.484 (0.003)	2558.3 (10.5)	2300.0 (10.7)
DYFAMED	2017-2022	74	12.913 (0.006)	38.485 (0.002)	2555.1 (11.8)	2297.3 (12.4)
ANTARES	2017-2023	62	12.944 (0.096)	38.485 (0.005)	2559.8 (9.0)	2302.2 (8.9)
MOOSE-GE	2017-2023	91	12.917 (0.005)	38.484 (0.003)	2559.8 (9.8)	2300.7 (10.0)
Tropical Pacific (layer 1800-2300m)						
OUTPACE	2017	15	2.124 (0.055)	34.633 (0.006)	2414.1 (8.0)	2318.8 (5.8)
TONGA	2019	7	2.196 (0.197)	34.619 (0.016)	2408.9 (9.1)	2327.2 (7.5)
Western Tropical Atlantic (1000m)						
AMAZOMIX	2021	14	4.770 (0.105)	34.711 (0.041)	2315.6 (20.2)	2220.8 (17.1)
TARA-MICRO	2021	1	4.852	34.717	2312.9	2231.1

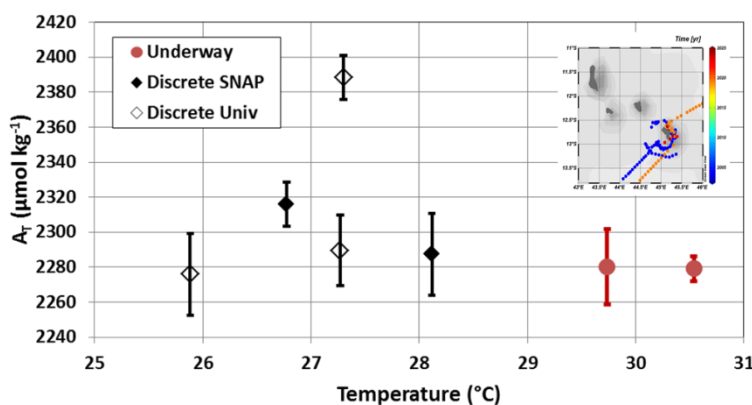
402 **Table 5:** Comparison of  $A_T$  and  $C_T$  analysed on-board and at SNAPO-CO2 facilities for the MINERVE project.  
403 The results are expressed as the standard deviations (Std) of the differences for each cruise. Nb = the number of  
404 co-located samples.

Period	Nb	Std $A_T$ $\mu\text{mol kg}^{-1}$	Std $C_T$ $\mu\text{mol kg}^{-1}$
2004-2005	109	12.85	4.99
2005-2006	45	4.20	6.77
2007-2008	17	10.15	10.62
2008-2009	26	15.80	12.02
2009-2010	22	4.04	5.78
2010-2011	33	9.36	6.83
2012-2013	29	5.43	9.73

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### 3.4.3 Comparison based on different techniques

Another example of comparison is presented for samples obtained in the lagoon of Mayotte Island in the western Indian Ocean and measured using different techniques. In the frame of the CARBODISS project seawater was sampled in 2018-2023 at several coral reef sites within the north-eastern part of the lagoon and measured either at LOCEAN laboratory or at La Réunion University. To remove coral sand particles the water samples were immediately filtered through Whatman GF/F filters and poisoned with mercuric chloride, following Dickson et al. (2007). In 2021, 2022 and 2023,  $A_T$  was measured at La Réunion University using an automated potentiometric titration (905 Titrandro Metrohm titrator with combined pH electrode 6.0253.00) and calculated from the second inflection point of the titration curve. The HCl concentration was checked each day of measurements using a CRM provided by A. Dickson, Scripps Institution of Oceanography. The  $A_T$  precision of  $\pm 2 \mu\text{mol kg}^{-1}$  was based on triplicate analyses (Lagoutte et al., 2023). In the studied coral reef sites  $A_T$  concentrations ranged between 2250 and 2350  $\mu\text{mol kg}^{-1}$  but with occasional higher concentrations up to 2450-2500  $\mu\text{mol kg}^{-1}$ . Such high  $A_T$  has been observed in other coral reefs ecosystems (Cyronak et al., 2013 at Cook Island; Palacio-Castro et al., 2023 at Middle Keys, Florida). The data obtained in the lagoon of Mayotte on different coral reefs could be compared with underway observations obtained offshore of Mayotte Island (OISO-11 cruises in 2004 and CLIM-EPARSES cruise in 2019, data available in the SNAPO-CO2-v1 dataset). In the open ocean the  $A_T$  concentrations ranged between 2250 and 2330  $\mu\text{mol kg}^{-1}$ , close to the results obtained at Mayotte reefs except for samples in November 2021 that were all collected at Cratère station (12.84°S-45.39°E) (Figure 4). At this location there was a large diurnal variation in November 2021 with  $A_T$  increasing from 2322 to 2508  $\mu\text{mol kg}^{-1}$  (Figure S5). This is because in 2021 the samples were taken at low tide recording a volcanic signal at this site allowing recording for the first time the volcanic signal in this location ( $\text{CO}_2$  resurgences). In 2018 and 2019 such high  $A_T$  were not measured (Figure S5) as samples were taken at high tides allowing a certain dilution of volcanic  $\text{CO}_2$  emissions in the water column. Although the samples were measured with different techniques the range of  $A_T$  is coherent for both datasets (Figure 4). Therefore we added the  $A_T$  data measured at La Réunion University in 2021-2023 to complete the synthesis for this location (Mayotte Island).



**Figure 4:** Total alkalinity ( $A_T$ ) versus temperature for samples measured around Mayotte and in the coral reef (insert map). Underway  $A_T$  was measured onboard in 2004 and 2019 (red circles) whereas discrete samples at different reef sites within the lagoon of Mayotte in 2018, 2019, 2021, 2022 and 2023 were measured at LOCEAN (black diamonds) or at La Réunion University (open diamonds). The figure presents the data averaged for each cruise in this region.

466 **3.4.4 Summary of quality control data**

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468 The total number of data in the SNAPO-CO2-v2 dataset for the Global Ocean is gathered in Table 6  
 469 with corresponding flags for each property. Overall, the synthesis includes more than 91% of good data for both  
 470  $A_T$  and  $C_T$ . About 6% are questionable and 3% are likely bad. Overall, we believe that all data (with flag 2) in  
 471 this synthesis have an accuracy better than  $4 \mu\text{mol kg}^{-1}$  for both  $A_T$  and  $C_T$ , the same as for quality-controlled  
 472 data in GLODAP (Lauvset et al., 2024). The uncertainty ranges between the ‘‘Climate goal’’ ( $2 \mu\text{mol kg}^{-1}$ ) and  
 473 the ‘‘Weather Goal’’ ( $10 \mu\text{mol kg}^{-1}$ ) for ocean acidification studies (Newton et al., 2015; Tilbrook et al., 2019).  
 474 This accuracy is also relevant to validate or constraint data-based methods that reconstruct  $A_T$  and  $C_T$  fields with  
 475 an error of around 10-15  $\mu\text{mol kg}^{-1}$  for both properties (Bittig et al., 2018; Broullón et al., 2019, 2020; Fourier et  
 476 al., 2020; Gregor and Gruber, 2021; Chau et al., 2024a).

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479 **Table 6:** Number of Temperature, Salinity,  $A_T$  and  $C_T$  data in the SNAPO-CO2-v2 synthesis identified for flags  
 480 2 (good), 3 (questionable), 4 (bad), 9 (no data). Last column is the percentage of flag 2 (Good).

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Property	Flag 2	Flag 3	Flag 4	Flag 9	% flag 2
Temperature	68253	418	0	653	99.4
Salinity	68706	482	5	131	99.3
$A_T$	61249	3910	2077	2088	91.1
$C_T$	61869	3865	2057	1533	91.3

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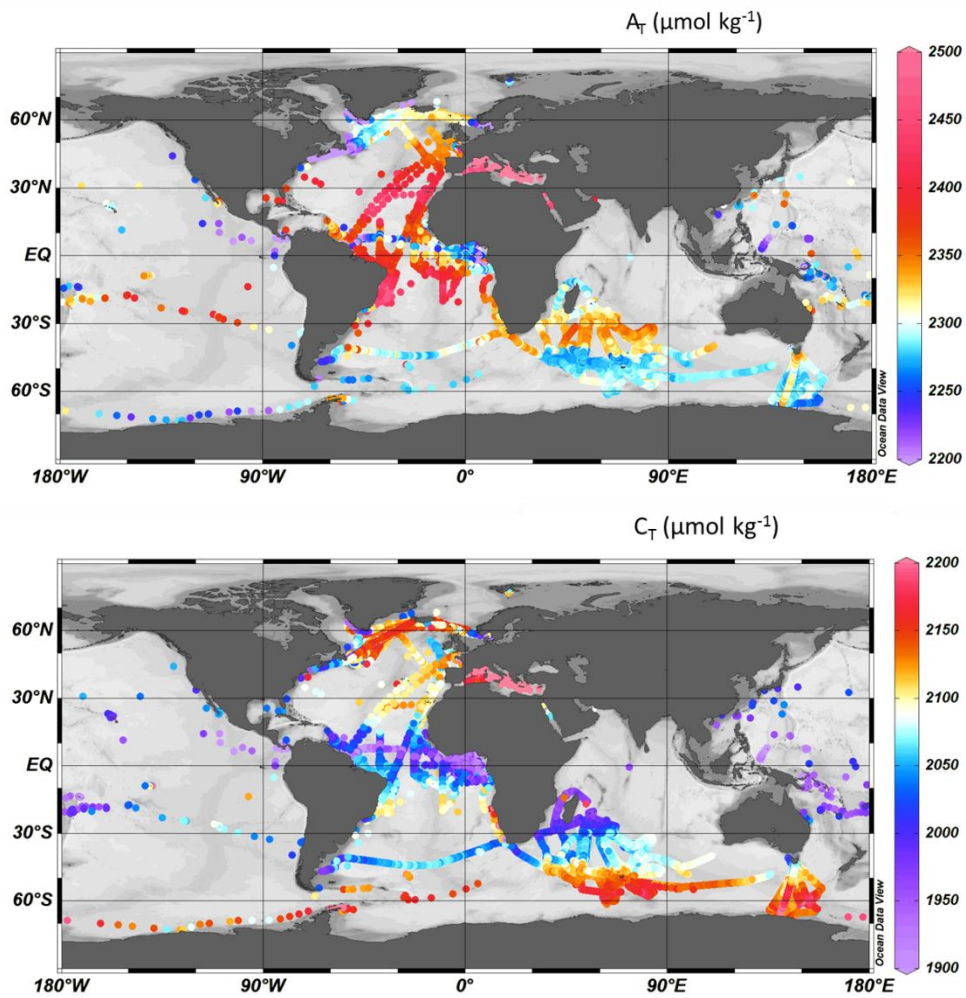
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492 **4 Global  $A_T$  and  $C_T$  distribution based on the SNAPO-CO2-v2 dataset**

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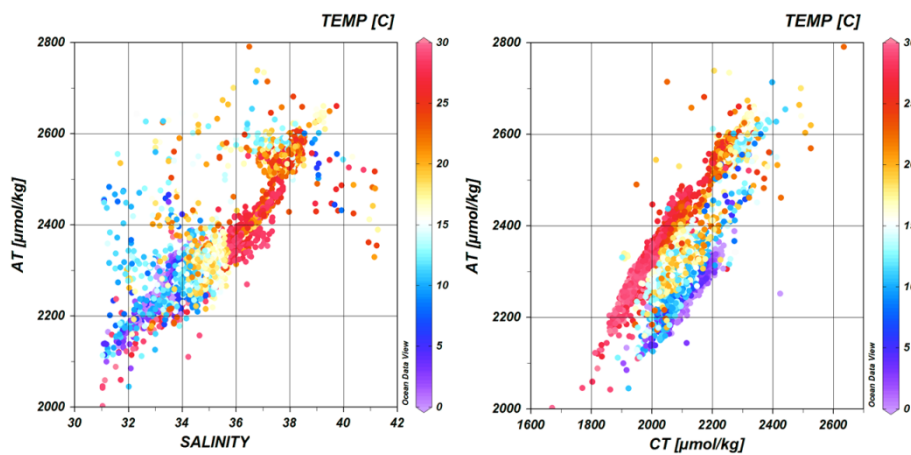
494 The surface distribution in the global ocean based on the SNAPO-CO2 dataset is presented in Figure 5  
 495 for  $A_T$  and  $C_T$ . The  $A_T$ /Salinity and  $A_T$ / $C_T$  relationships are clearly identified and structured at regional scale  
 496 (Figure 6). In the open ocean, high  $A_T$  concentrations ( $> 2400 \mu\text{mol kg}^{-1}$ ) are identified in the Atlantic subtropics  
 497 (bands  $35^\circ\text{N}$ - $15^\circ\text{N}$  and  $25^\circ\text{S}$ - $3^\circ\text{S}$ ) (Jiang et al., 2014; Takahashi et al., 2014). The lowest  $A_T$  and  $C_T$   
 498 concentrations ( $< 600 \mu\text{mol kg}^{-1}$ ) are observed in the western tropical Atlantic in the Amazon River plume near  
 499 the mouth (Lefèvre et al., 2017b). For  $C_T$  the concentrations are high ( $> 2150 \mu\text{mol kg}^{-1}$ ) in the Southern Ocean  
 500 south of the polar front, associated with the deep mixing in winter and the upwelling of deep water (Metzl et al.,  
 501 2006; Pardo et al., 2017). The highest  $C_T$  concentrations (up to  $2180$ - $2270 \mu\text{mol kg}^{-1}$ ) are observed in the high  
 502 latitudes of the Southern Ocean near the Adélie coastal zone (MINERVE and ACE cruises), around the  
 503 Kerguelen plateau (OISO-31 cruise) and close to the Antarctic Peninsula (TARA-Microbiome cruise). In the  
 504 North Atlantic the new data from SURATLANT cruises in 2018-2023 confirm the high  $C_T$  concentrations ( $>$   
 505  $2150 \mu\text{mol kg}^{-1}$ ) observed in the Sub-polar gyre since 2016 due in part to the accumulation of anthropogenic  $\text{CO}_2$   
 506 (Leseurre et al., 2020). Low  $C_T$  concentration ( $< 2000 \mu\text{mol kg}^{-1}$ ) are found in the tropics ( $10^\circ\text{N}$ - $30^\circ\text{S}$ ) with  
 507 lower values ( $< 1950 \mu\text{mol kg}^{-1}$ ) in the equatorial Atlantic band  $10^\circ\text{N}$ -Eq (e.g. Koffi et al., 2010; Lefèvre et al.,  
 508 2021). In the Amazon shelf sector  $C_T$  can reach even lower concentration ( $< 1700 \mu\text{mol kg}^{-1}$ , AMAZOMIX  
 509 cruise).

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**Figure 5:** Distribution of  $A_T$  (top) and  $C_T$  (bottom) concentrations ( $\mu\text{mol.kg}^{-1}$ ) in surface waters (0-10m) in the SNAPO-CO2-v2 dataset. Only data with flag 2 are presented in these figures. Figures produced with ODV (Schlitzer, 2018).



**Figure 6:** Relationships between  $A_T$  and Salinity (left panel) and  $A_T$  versus  $C_T$  (right panel) for samples in surface waters (0-10m and Salinity > 31). Only data with flag 2 are presented (nb = 48749). The color scales correspond to the temperature. The data not aligned correspond to coastal zones (e.g. COCORICO2 stations). Figures produced with ODV (Schlitzer, 2018).

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## 5 Regional $A_T$ and $C_T$ distributions and trends based on the SNAPO-CO2 dataset

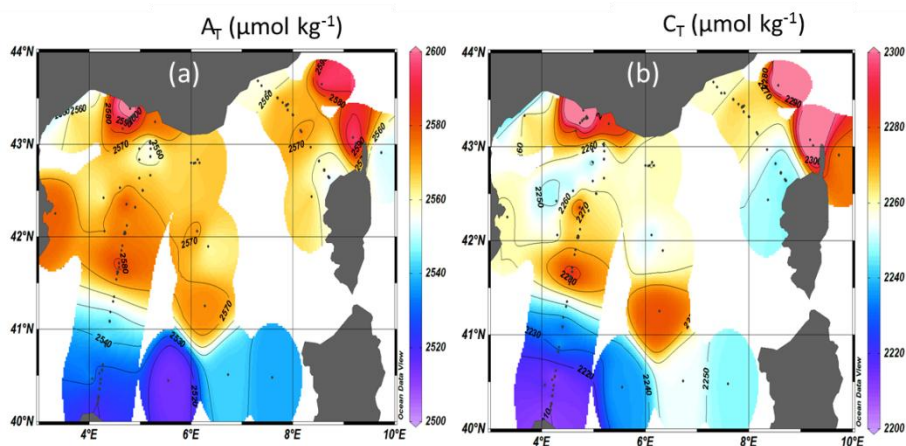
The regional distributions are described for the Mediterranean Sea and for selected regions in the open ocean and coastal zones where the data are available for 10 years or more to explore the  $A_T$  and  $C_T$  trends. Given the observed seasonal and inter-annual variability and that the time-series were not regular (e.g. at monthly frequency), we cannot use recommended methods to estimate the trends (e.g. based on de-seasoned data, Sutton et al, 2022). Here we have selected the locations and seasons where the  $C_T$  trends can be linearly fitted and compared with no interpolation to fill gaps and discontinuous data (e.g., fewer samples during the COVID period).

### 5.1 The Mediterranean Sea

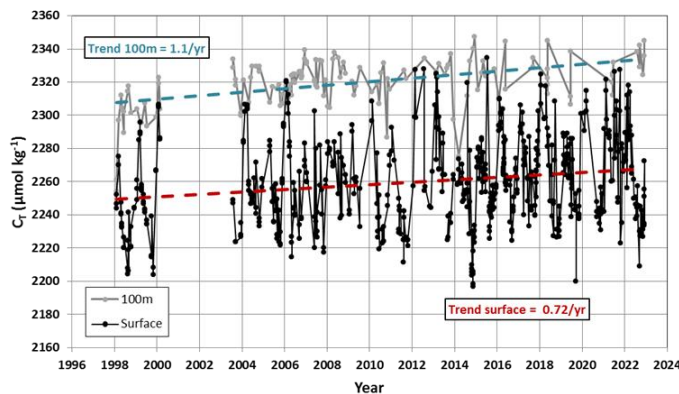
Compared to the open ocean,  $A_T$  concentrations are much higher in the Mediterranean Sea (Copin-Montégut, 1993; Schneider et al., 2007; Álvarez et al., 2023) with values up to 2600  $\mu\text{mol kg}^{-1}$ . The  $A_T$  and  $C_T$  data obtained in 2014-2023 show a clear contrast between the northern and southern regions of the Western Mediterranean Sea with higher concentration in the Ligurian Sea and the Gulf of Lion (Figure 7). This contrast is associated to the circulation and the frontal system in this region (e.g. Barral et al, 2021). New data in the coastal zones in the Gulf of Lion (ACCESS, DICASE, CARBODELTA, COCORICO2, MESURHOBENT) also have very high  $A_T$  and  $C_T$  concentrations ( $A_T > 2600 \mu\text{mol kg}^{-1}$ ;  $C_T > 2350 \mu\text{mol kg}^{-1}$ ). Very low  $A_T$  and  $C_T$  concentrations ( $A_T < 2500 \mu\text{mol kg}^{-1}$ ;  $C_T < 2200 \mu\text{mol kg}^{-1}$ ) were also occasionally observed in the coastal zones (COCORICO2 stations, Petton et al, 2024).

In summer 2022 the Mediterranean Sea experienced an exceptional warming (Figure S6) superposed to the long-term warming in the ocean (Cheng et al, 2024). Such event would impact the internal ocean processes such as thermodynamic, stratification and biological processes (Coppola et al., 2023) and the inter-annual variability and trends of  $C_T$ , pH,  $f\text{CO}_2$  and air-sea  $\text{CO}_2$  fluxes (Yao et al., 2016; Wimart-Rousseau et al., 2023; Chau et al., 2024b). As in 2003, the warming in summer 2022 was associated to the drought event that occurred in Europe and over the Mediterranean Sea (Faranda et al., 2023). In July 2022, the maximum temperature of 28.42°C was observed at station SOMLIT-Point-B. In the Ligurian Sea the temperature trend was faster in recent years,  $+0.173 \pm 0.072 \text{ }^\circ\text{C per decade over 1990-2010}$  and  $+0.678 \pm 0.143 \text{ per decade over 2010-2023}$  (Figure S6). With the new data added in the SNAPO-CO2-v2 synthesis (DYFAMED, MOOSE-ANTARES, and MOOSE-GE) we evaluated a temperature trend of  $+0.84 \pm 0.20 \text{ }^\circ\text{C per decade over 1998-2022}$  indicating that the discrete sampling captured the property changes at regional scale. Based on the data in the Ligurian Sea the trends of  $C_T$  appeared faster in summer ( $+1.53 \pm 0.46 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ ) than in winter ( $+0.94 \pm 0.64 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ , Table 7). On the other hand, the trends of  $A_T$  were the same ( $+0.72 \pm 0.36 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in winter and  $+0.69 \pm 0.42 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in summer). The trend of  $C_T$  in surface in winter was close to the one derived at 100m (below the Chl-a maximum),  $C_T^{100\text{m}} = +1.10 \pm 0.17 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  (Figure 8) whereas for  $A_T$  the trend was the same in surface and at depth ( $+0.76 \pm 0.12 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ ). This suggests that the winter  $C_T$  data recorded the anthropogenic  $\text{CO}_2$  uptake of around  $+1 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ , Figure S7). Note that, given the observed  $C_T$  trends, the spatial view presented in Figure 7b for 2014-2023 would be the same based on  $C_T$  concentrations normalized to a reference year. As noted by Touratier and Goyet (2009) the  $C_T$  concentrations in the Mediterranean Sea should

605 increase in parallel with the level of atmospheric anthropogenic CO<sub>2</sub>. For an atmospheric CO<sub>2</sub> rate of +2.16 ppm  
 606 yr<sup>-1</sup> over 1998-2023 (Lan et al., 2024) and at fixed sea surface temperature (17.75°C), salinity (38.25) and A<sub>T</sub>  
 607 (2567 μmol kg<sup>-1</sup>), the theoretical C<sub>T</sub> increase would be +1.24 μmol kg<sup>-1</sup> yr<sup>-1</sup>. Interestingly, an anthropogenic flux  
 608 of -0.3 ± 0.02 molC m<sup>-2</sup> yr<sup>-1</sup> in the Mediterranean Sea (Bourgeois et al., 2016) would correspond to an increase of  
 609 C<sub>T</sub> of 1.07 ± 0.07 μmol kg<sup>-1</sup> yr<sup>-1</sup> in the top 100 meters. This is again close to what is observed in winter or at  
 610 100m (Table 7, Figure 8). On the other hand the faster C<sub>T</sub> trend observed in surface waters during summer might  
 611 be associated with a decrease in biological production and/or changes in circulation/mixing over time that  
 612 deserve specific investigations such as analyzed for the oxygen budget in this region (Ulses et al, 2021). It is  
 613 worth noting that the C<sub>T</sub> and A<sub>T</sub> trends in coastal zones of the Mediterranean Sea are opposite to those observed  
 614 offshore: for example at station SOLEMIO (Bay of Marseille, Wimart-Rousseau et al., 2020) the C<sub>T</sub> and A<sub>T</sub>  
 615 concentrations decreased over 2016-2022 and thus opposed to the anthropogenic CO<sub>2</sub> signal, indicating that  
 616 processes such as riverine inputs, advection or biology control the carbonate system decadal variability at local  
 617 scale. This calls for developing dedicated complex biogeochemical models to resolve these processes (Barré et  
 618 al., 2023, 2024), especially when extreme events occurred, such as the very hot summer in 2024 with SST up to  
 619 30°C in the Mediterranean Sea (Platforms Buoy/Mooring AZUR, EOL and La Revellata, data available at  
 620 <https://dataselection.coriolis.eu.org/>). The data obtained in the Mediterranean Sea are important not only to  
 621 validate biogeochemical models but also to reconstruct the carbonate system from A<sub>T</sub> and pCO<sub>2</sub> data (Chau et al.,  
 622 2024a) as the global A<sub>T</sub>/SSS relationships (e.g. Carter et al., 2018) are not suitable for this region.



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**Figure 7:** Distribution of A<sub>T</sub> (a) and C<sub>T</sub> (b) in μmol kg<sup>-1</sup> in surface waters of the Mediterranean Sea (0-10m) from  
 634 observations over 2014-2023. Figures produced with ODV (Schlitzer, 2018).  
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652 Figure 8: Time-series of  $C_T$  concentrations in surface (black symbols) and at 100m (grey symbols) in the  
653 Ligurian Sea. The trends over 1998-2022 is surface (red) and at 100m (blue) are indicated by dashed lines.  
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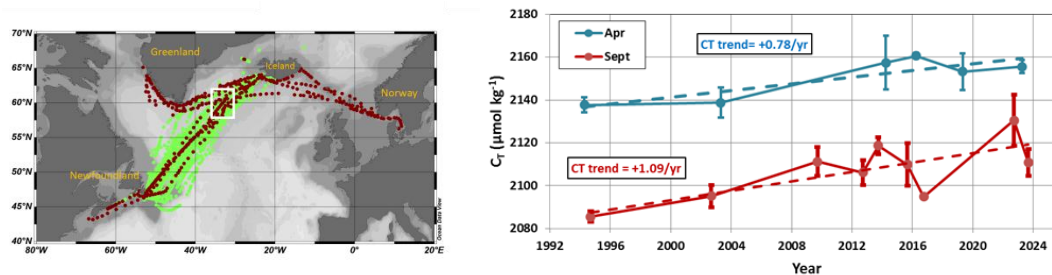
## 655 5.2 The North Atlantic

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657 The North Atlantic Ocean is an important  $\text{CO}_2$  sink (Takahashi et al. 2009) due to biological activity  
658 during summer, heat loss and deep convection during winter. As a result this region contains high concentrations  
659 of anthropogenic  $\text{CO}_2$  ( $C_{\text{ant}}$ ) in the water column (Khatiwala et al., 2013). Decadal variations of the  $C_{\text{ant}}$   
660 inventories were recently identified at basin scale probably linked to the change of the overturning circulation  
661 (Gruber et al., 2019; Müller et al., 2023; Pérez et al., 2024). This region experienced climate modes such as the  
662 North Atlantic Oscillation (NAO) and the Atlantic Multidecadal Variability (AMV) that imprint variability in  
663 air-sea  $\text{CO}_2$  fluxes at inter-annual to multidecadal scales (e.g. Thomas et al., 2008; Jing et al., 2019;  
664 Landschützer et al., 2019) but not always clearly revealed at regional scale (Metzl et al., 2010; Schuster et al.,  
665 2013; Pérez et al., 2024). In addition it has been recently shown that extreme events such as the marine heat  
666 wave in summer 2023 led to a reduce  $\text{CO}_2$  uptake in this region (Chau et al., 2024b). Although the annual  
667  $\text{CO}_2$  fluxes deduced from Global Ocean Biogeochemical Models (GOBM) seem coherent with the data-products  
668 at basin scale (resp.  $-0.30 \pm 0.07$  and  $-0.24 \pm 0.03$  PgC/yr for the North Atlantic subpolar seasonally stratified,  
669 NA-SPSS biome) the  $p\text{CO}_2$  cycle seasonality is not well simulated (Pérez et al., 2024). Therefore to correct the  
670 GOBMs outputs, comparisons with the observed  $C_T$  and  $A_T$  cycles are also needed.

671 In this context regular sampling in the North Atlantic (OVIDE cruises, Mercier et al., 2015, 2024;  
672 SURATLANT transects, Reverdin et al., 2018) and time-series stations in the Irminger and Iceland Seas  
673 (Ólafsson, et al., 2010; Lange et al., 2024; Yoder et al., 2024) are important to explore the variability of the  
674 biogeochemical properties from seasonal (Figure S8) to decadal scales (Figure 9). The SURATLANT data added  
675 in the SNAPO-CO2-v2 dataset over 2017-2023 offer new observations in the North Atlantic Subpolar Gyre  
676 (NASPG in the NA-SPSS biome) and new transects from Norway to Iceland and reaching the coast of Greenland  
677 (Figure 9). In 2010 the winter NAO was negative, moved to a positive state in 2012-2020 and was again very  
678 low in 2021. The new SURATLANT data after 2017 confirm the cooling and the freshening in the NASPG since  
679 2009 (Holliday et al., 2020; Leseurre et al., 2020; Siddiqui et al., 2024) whereas the most recent data in 2022 and  
680 2023 suggest a reverse trend (increase of salinity and temperature, Figure S8). After 2016, large  $C_T$  anomalies in  
681 the NASPG were observed. For examples, in April 2019 and 2022, the  $C_T$  concentrations were low compared to  
682 2016 (Figure 9) and opposed to the expected anthropogenic  $\text{CO}_2$  uptake. In September 2023 the  $C_T$   
683 concentrations were much lower than in 2022 (Figure 9) probably linked to biological productivity when the  
684 NAO index was negative (Fröb et al., 2019) as observed in summer 2023 (NAO < -2 in July 2023). Despite these  
685 variability the  $C_T$  trends are relatively well evaluated (Table 7). As in the Mediterranean Sea the  $C_T$  trends in the  
686 NASPG appeared different depending on the season (Figure 9). The  $C_T$  increase was faster in September than in  
687 April (resp.  $+1.09 \pm 0.37 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  and  $+0.78 \pm 0.23 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ). This is either close to or lower than the  
688 theoretical  $C_T$  increase due to the rising of atmospheric  $\text{CO}_2$  ( $+0.91 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ) and in the range of recent  
689 results evaluated for the Sub-polar Mode Waters in the Irminger Sea ( $C_{\text{ant}}$  trend =  $0.95 \pm 0.17 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  for  
690 the period 2009-2019, Curbelo-Hernández et al., 2024).

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**Figure 9:** Left: Data in SNAPO-CO2-v1 (green) and new data in v2 (brown) from the SURATLANT cruises in 1993-2023 in the North Atlantic. Figure produced with ODV (Schlitzer, 2018). The white box identified the region of selected data around 60°N for the trend analysis. Right: Time-series of average  $C_T$  concentrations in April (blue) and September (red) in this region. The trends for each season are indicated (see also Table 7).

### 707 5.3 The Tropical Atlantic

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709 In the Tropical Atlantic, previous studies highlighted the large variability of biogeochemistry and the  
710 difficulty in detecting long-term trends of  $C_T$  (e.g. Lefèvre et al., 2021). This is related to the variability of  
711 circulation, equatorial upwelling, biological processes (some linked to Saharan dust) and inputs from large rivers  
712 (Congo, Amazon and Orinoco). The new data added in version SNAPO-CO2-v2 (Figure S9) show the  
713 contrasting zonal  $C_T$  distribution in this region with lower concentrations in low salinity regions of the North  
714 Equatorial Counter Current and Guinea Current (Figure 5; Oudot et al., 1995; Takahashi et al., 2014; Broullón et  
715 al., 2020; Bonou et al., 2022). For exploring the temporal changes we selected the data in the western region  
716 available for at least 10 years and separated the northern and southern sectors. In both regions the  $C_T$  trend is  
717 close to  $+3 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  (Table 7, Figure S9) much higher than the expected anthropogenic signal. In this  
718 region where coastal water masses mixes with oceanic waters, the inter-annual variability of  $C_T$  is large and the  
719 changes driven by competitive processes (circulation, biological processes). More observations and dedicated  
720 models are needed to separate the anthropogenic and natural variability in this region (Pérez et al., 2024).

### 721 722 5.4 The Southern Ocean

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724 In the Southern Ocean there are a few regular multi-annual observations of the carbonate system. Time  
725 series of more than 10 years were obtained in the Drake Passage (Munro et al., 2015) and in the Southern Indian  
726 Ocean (Leseurre et al., 2022; Metzl et al., 2024b). Observations were also obtained for more than 20 years  
727 southeast of New Zealand at the Munida Time Series (MTS) in the subtropical and sub-Antarctic frontal zones  
728 (Currie et al., 2011; Vance et al., 2024). To complement these datasets we have added the data collected in the  
729 South-Eastern Indian Ocean between Tasmania and Antarctica in the frame of the MINERVE cruises (Figure 10;  
730 Brandon et al., 2022). These cruises were conducted from October to March offering each year a view of the  
731 seasonal changes between late winter and summer from the sub-Antarctic zone to the coastal zone near  
732 Antarctica (Adélie land). In all sectors (here from 45°S to 67°S) the  $C_T$  concentrations were higher in October  
733 when the mixed-layer depth (MLD) was deep and were lower during the productive summer season (e.g. Laika  
734 et al., 2009; Shadwick et al., 2015). An example is presented at 60°S/151°E from the data obtained along a  
735 reoccupied track in 2011-2012 (Figure S10). At this location south of the Polar Front in the POOZ/HNLC area



736 (Permanent Open Ocean Zone/ High Nutrient Low Chlorophyll), the  $C_T$  concentrations were  $+25 \mu\text{mol kg}^{-1}$   
 737 higher in October compared to February. The same seasonal amplitude was observed in the western Indian sector  
 738 of the POOZ (Metzl et al., 2006, 2024b) suggesting that the  $C_T$  seasonality is relatively homogeneous in this  
 739 region corresponding to the Indian SO-SPSS biome (Fay and McKinley, 2014). The difference in the  
 740 climatological  $C_T$  between October and January is on average  $+28.3 \pm 9.8 \mu\text{mol kg}^{-1}$  in the Indian Ocean POOZ  
 741 (Takahashi et al., 2014). Given this seasonality and potential change in the seasonal amplitude over time  
 742 (Gallego et al., 2018; Landschützer et al., 2018; Shadwick et al., 2023) the property trends have to be evaluated  
 743 for October and January-February separately, here over 2002-2012 in the POOZ (Figure 10, Table 7). In both  
 744 seasons, the average  $C_T$  concentrations reached a minimum in 2008 and increased faster in 2008-2012 (up to  
 745  $+4.8 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ). Interestingly, such acceleration of the trend after 2009 was observed for  $p\text{CO}_2$  at the MTS  
 746 station (Vance et al., 2024). We note that the  $C_T$  trend over 2002-2012 was slightly faster in October (Figure 10)  
 747 probably linked to deeper MLD as suggested from the cooling and the salinity increase observed during this  
 748 season (Figure S10).

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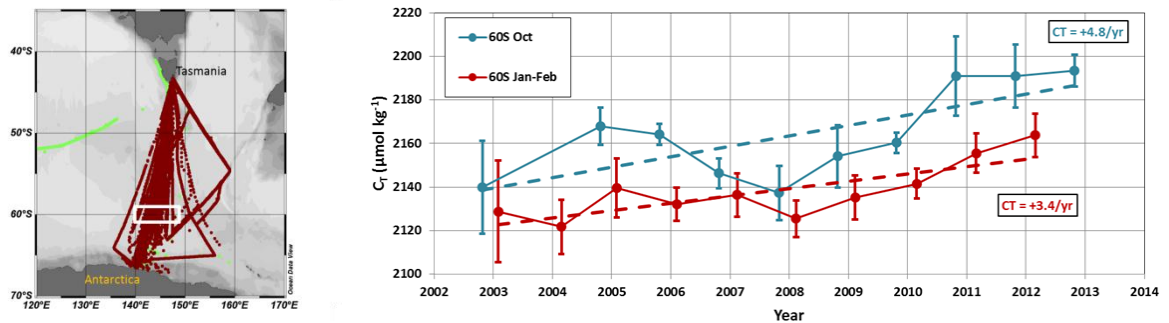
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761 **Figure 10:** Left: Data in SNAPO-CO2-v1 dataset (green) and new data in version v2 (brown) in the South  
 762 eastern Indian Ocean. Figure produced with ODV (Schlitzer, 2018). The white box identified the region of  
 763 selected data around 60°S for the trend analysis. Right: Time-series of average  $C_T$  concentrations in January-  
 764 February (red) and October (blue) around 60°S (white box in the map). The trends for each season are indicated  
 765 (see also Table 7).

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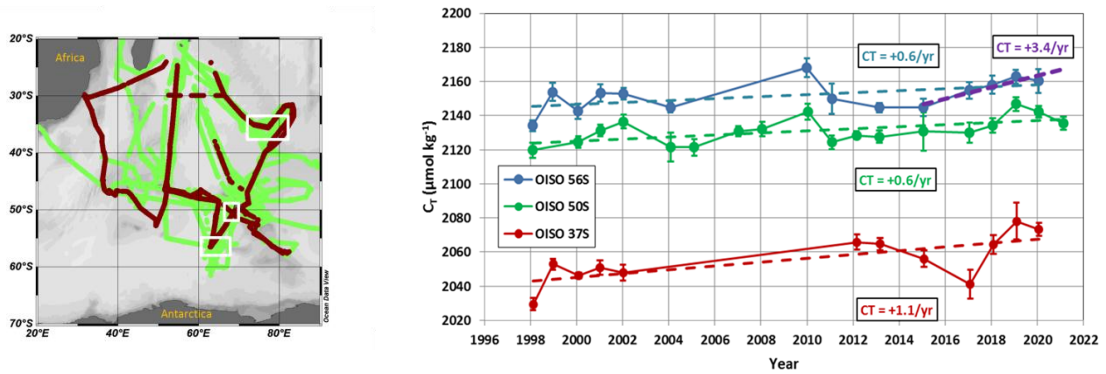
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In the western Indian sector, the new data in the SNAPO-CO2-v2 dataset from the OISO cruises at high  
 latitudes also recorded a rapid  $C_T$  trend over 5-8 years periods (e.g.,  $+3.4 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  in 2015-2020 at 56°S,  
 Figure 11, Table 7). Although the inter-annual variability of  $C_T$ , between 10 and 20  $\mu\text{mol kg}^{-1}$ , is often  
 recognized (Figure 11), the evaluation of the trends over more than 20 years indicated faster trend in the  
 subtropical Indian Ocean ( $+1.1 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ) compared to higher latitudes (Indian POOZ,  $+0.6 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ );  
 they are close to the expected anthropogenic signal in these regions ( $+1.1 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  in the subtropics and  
 $+0.8 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  at higher latitudes).

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**Figure 11:** Left: Data in SNAPO-CO2-v1 dataset (green) and new data in version v2 (brown) in the South Western Indian Ocean (OISO cruises). Figure produced with ODV (Schlitzer, 2018). The white boxes identified the regions of data selected around 37°S, 50°S and 56°S for the trend analysis. Right: Time-series of average  $C_T$  concentrations in January-February at 37°S (red), 50°S (green) and 56°S (blue). The trends for each region are indicated (see also Table 7).

**Table 7:** Trend of  $C_T$  ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ ) and corresponding standard error in selected regions where data are available for more than 10 years. The projects/cruises for selection of the data in each domain are indicated.

Region	Period/Season	$C_T$ trend ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	Projects/Cruises
North Atlantic (NASPG)	1994-2023 April	+0.78 (0.23)	SURATLANT
North Atlantic (NASPG)	1994-2023 September	+1.09 (0.37)	SURATLANT
West. Trop. Atl. 5N-Eq	2009-2021 April-October	+3.31 (2.13)	AMAZOMIX, PIRATA-BR, TARA
West. Trop. Atl. Eq-10S	2005-2015, April-October	+3.05 (1.64)	CAMFIN-WAT, PIRATA-BR, VOS
Ligurian Sea 8E	1998-2022 Jan-Feb.	+0.94 (0.64)	ANTARES, DYFAMED, MOOSE-GE
Ligurian Sea 8E	1998-2023 July-August	+1.53 (0.46)	ANTARES, DYFAMED, MOOSE-GE
Subtropical Indian 37S	1998-2020 Jan-Feb.	+1.12 (0.36)	OISO
South West. Indian 50S	1998-2021 Jan-Feb.	+0.61 (0.21)	OISO
South West. Indian 56S	1998-2020 Jan-Feb.	+0.58 (0.27)	OISO
South West. Indian 56S	2015-2020 Jan-Feb.	+3.41 (0.73)	OISO
South East. Indian 60S	2002-2012 Jan-Feb.	+3.37 (0.94)	MINERVE, OISO
South East. Indian 60S	2002-2012 October	+4.79 (1.62)	MINERVE

## 824 5.5 The Coastal Zones

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826 Coastal waters experience enhanced ocean acidification due to increasing  $\text{CO}_2$  uptake, accumulation of  
827 anthropogenic  $\text{CO}_2$  (Bourgeois et al 2016; Laruelle et al, 2018; Roobaert et al, 2024a; Li et al, 2024) and from  
828 local anthropogenic inputs through rivers or from air pollution (e.g. Sarma et al, 2015; Sridvi and Sarma, 2021;  
829 Wimart-Rousseau et al, 2020). The changes of the  $\text{CO}_2$  uptake in coastal zones are also linked to biological  
830 processes (Mathis et al, 2024) or to circulation and local upwelling (Roobaert et al, 2024b), all controlling large  
831 variability of  $A_T$  and  $C_T$  in space and time leading to uncertainties for detecting long-term changes of  $p\text{CO}_2$  and  
832 air-sea  $\text{CO}_2$  fluxes in heterogeneous coastal waters (Dai et al 2022; Resplandy et al, 2024). At seasonal scale,  
833 large differences between observations and models were also identified leading to differences in the coastal

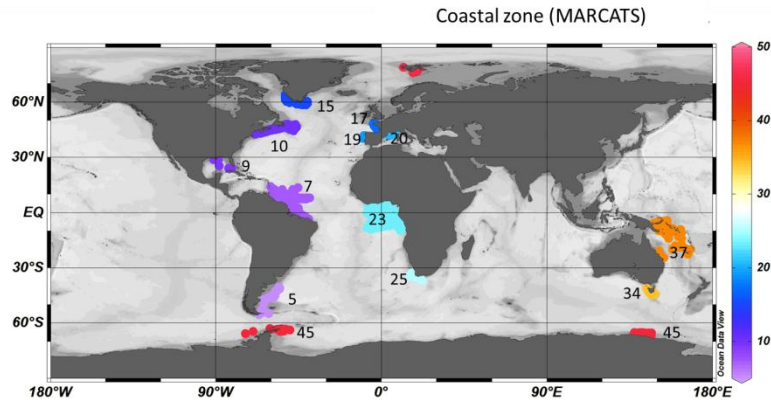
834 ocean CO<sub>2</sub> sink up to 60% (Resplandy et al, 2024). It is thus important to document the seasonal cycles of A<sub>T</sub> and  
835 C<sub>T</sub> to compare and correct models and thus to better predict future changes of biogeochemical properties in  
836 coastal waters and their impact on marine ecosystems. A better understanding of the processes and their  
837 retroaction in the coastal regions is also required regarding Marine Carbon Dioxide Removal (MCDR)  
838 experiments and for their evaluation (e.g. Ho et al, 2023).

839 In the SNAPO-CO<sub>2</sub>-v2 dataset new data have been added in the coastal zones at stations SOMLIT-  
840 Brest, SOMLIT-Roscoff and SOMLIT-Point-B. They extend the period to 2022 or 2023 for temporal analysis.  
841 New data in the French coastal zones have been also included from the COCORICO2 project documented in  
842 detail by Petton et al (2024). The observations in coastal zones could be identified in the MARCATS regions  
843 (Margins and CATchment Segmentation, Laruelle et al, 2013) (Figure 12) where little information is available  
844 for quantifying the ocean CO<sub>2</sub> sink at the decadal scale and for evaluation of the anthropogenic CO<sub>2</sub> uptake  
845 (Regnier et al, 2013; Dai et al, 2022; Li et al, 2024). To explore the change of the observed properties in the  
846 coastal zones and have a flavor of the long-term C<sub>T</sub> trends we selected the time series with at least 10 years of  
847 data (Table 8, Figure 13). Except at high latitudes (Greenland and Antarctic coastal zones), we observed a  
848 warming in coastal zones (Figure S11). Changes in salinity are also identified (increase or decrease) and results  
849 of the trends are presented for salinity-normalized C<sub>T</sub> at 34, 35 or 38 depending on the region. Although the  
850 inter-annual variability is large in coastal waters, sometimes linked to extreme events (e.g. river discharges), we  
851 observed an increase in N-C<sub>T</sub> at most of the 8 selected locations. The exceptions are the coastal zones in the Gulf  
852 of Lion near the Rhone River and near Tasmania in October.

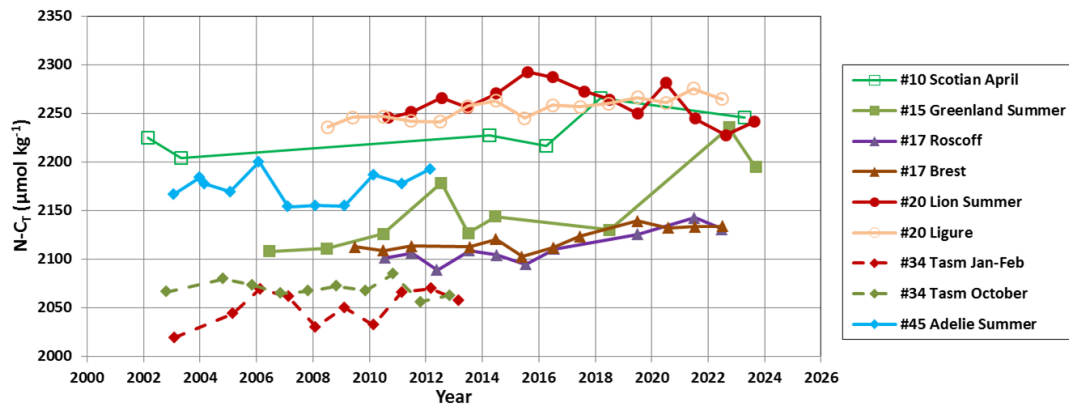
853 In the Gulf of Lion, the new data in the coastal zone confirmed the first view at the SOLEMIO station  
854 over 2016-2018 (Bay of Marseille, Wimart-Rousseau et al, 2020). In this region the lowest C<sub>T</sub> was observed in  
855 summer 2022 (average C<sub>T</sub> of 2238.6 ±21.0 μmol kg<sup>-1</sup>), much lower than in 2015 (2290.8 ±44.7 μmol kg<sup>-1</sup>). Over  
856 the continental shelf south of Tasmania (MARCATS #34), the trend in N-C<sub>T</sub> was positive in summer but not  
857 significant in October. In October this was associated with an increase in Salinity and in A<sub>T</sub> probably linked to  
858 advective processes via the reversal and variability of the Zeehan or the East Australian currents. From our data a  
859 warming of +0.06°C yr<sup>-1</sup> was identified for both seasons over 2002-2012 as previously observed south of  
860 Tasmania over 1991-2003 impacting the pCO<sub>2</sub> trend and air-sea CO<sub>2</sub> fluxes in this region (Borges et al, 2008).  
861 The difference in the N-C<sub>T</sub> trends in austral summer and spring calls for new detailed studies with extended data  
862 in this region. At high latitude in the Adélie Land (Antarctic coast MARCATS #45), the variability of N-C<sub>T</sub> was  
863 large (range from 2150 to 2200 μmol kg<sup>-1</sup>, Figure 13) and the trend over 10 years in summer was not significant  
864 (Table 8). As opposed to the open zone at 60°S (Figure 10) the C<sub>T</sub> concentrations in the coastal zone near  
865 Antarctica were not increasing, probably linked to competitive processes between anthropogenic uptake, changes  
866 in primary production, mixing or ice melting (Shadwick et al, 2013, 2014). More data are needed to better  
867 evaluate the changes of the carbonate system in Antarctic coastal zones where bottom waters are formed and  
868 transport anthropogenic CO<sub>2</sub> at lower latitudes (Zhang et al, 2023).

869 For the coastal time series SOMLIT where annual trends could be estimated (sampling at monthly  
870 resolution), the N-C<sub>T</sub> increase (+2.1 to 3.4 μmol kg<sup>-1</sup> yr<sup>-1</sup>) is close or higher than the anthropogenic signal leading  
871 to a decrease in pH ranging between -0.05 to -0.06 TS decade<sup>-1</sup>. The new data added in the SNAPO-CO<sub>2</sub>-v2  
872 dataset (2016-2023) confirm the progressive increase in C<sub>T</sub> and the acidification in the western Mediterranean  
873 Sea and in the North-East Atlantic coastal zones (Kapsenberg et al, 2017; Gac et al, 2021).

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**Figure 12:** Location of  $A_T$  and  $C_T$  data available in the coastal zones in the SNAPO-CO2-v2 dataset. Numbers and Color code identify MARCATS region (Laruelle et al, 2013). Figure produced with ODV (Schlitzer, 2018).



**Figure 13:** Time-series of average  $N-C_T$  concentrations ( $\mu\text{mol kg}^{-1}$ ) in selected MARCATS regions for different period when data are available for ten years or more. The trends and periods for each region are indicated in Table 8.

**Table 8:** Trends of  $N-C_T$  ( $\mu\text{mol kg}^{-1} \text{ yr}^{-1}$ ) and corresponding standard errors in selected coastal regions where data are available for 10 years or more. The projects/cruises for selection of the data in each domain are indicated. MARCATS # regions also identified. Salinity values used for  $C_T$  normalization are indicated.

Region #MARCATS	Period	Season	$N-C_T$ Trend ( $\mu\text{mol kg}^{-1} \text{ yr}^{-1}$ )	Salinity	Projects/Cruises
Scotian #10	2002-2023	March-April	+1.71 (0.97)	35	SURATLANT
Greenland #15	2006-2023	June-mid-Sept	+5.77 (1.62)	35	OVIDE, SURATLANT
Roscoff #17	2010-2022	All season	+3.40 (0.76)	35	CHANNEL, COCORICO2, SOMLIT ROSCOFF
Bay of Brest #17	2009-2022	All seasons	+2.17 (0.52)	35	SOMLIT-Brest, COCORICO2, ECOSCOPIA,
LION#20	2010-2023	June-Sept	-1.19 (1.25)	38	COCORICO2, MOOSE-GE, SOLEMIO (a)
LIGURE#20	2008-2022	All seasons	+2.12 (0.36)	38	SOMLIT-Point-B, MOOSE-GE
Tasmania #34	2003-2013	Jan-Feb	+2.73 (1.72)	35	MINERVE, OISO
Tasmania #34	2002-2012	Oct	-0.65 (0.89)	35	MINERVE, OISO
Adélie #45	2002-2012	Dec-Feb	+0.63 (0.70)	34	MINERVE, OISO

(a) For LION, some data in summer were also used from punctual cruises: AMOR-BFlux, CARBORHONE, DICASE, LATEX, MESURHOBENT, MISSRHODIA2 and MOLA.

## 934 6 Summary and suggestions

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936 This work extends in time and in new oceanic regions the  $A_T$  and  $C_T$  data presented in the first SNAPO-  
937 CO<sub>2</sub> synthesis (Metzl et al, 2024a). It includes now more than 67 000 surface and water column observations in  
938 all oceanic basins, in the Mediterranean Sea, in the coastal zones, near coral reefs, and in rivers. The data  
939 synthesized in version v2 are based on measurements of  $A_T$  and  $C_T$  performed between 1993 and 2023 with an  
940 accuracy of  $\pm 4 \mu\text{mol kg}^{-1}$ . Based on a secondary quality control, 91% of the  $A_T$  and  $C_T$  data are considered as  
941 good (WOCE Flag 2) and 6% probably good (Flag 3). For the open ocean this synthesis complements the  
942 SOCAT, GLODAP and SPOTS data products (Bakker et al., 2016; Lauvset et al., 2024; Lange et al, 2024). For  
943 the coastal sites this also complements the synthesis of coastal time-series in the Iberian Peninsula (Padin et al,  
944 2020), in the Canadian Atlantic continental shelf (Gibb et al, 2023) and around North America (Fassbender et al.,  
945 2018; Jiang et al., 2021; Jiang et al 2024, in prep). The SNAPO-CO<sub>2</sub> dataset enables to investigate the seasonal  
946 cycles, the inter-annual variability and the decadal trends of  $A_T$  and  $C_T$  in various oceanic provinces. The same  
947 temporal analyses could be investigated for other carbonate system properties such as  $f\text{CO}_2$  or pH calculated  
948 from  $A_T$  and  $C_T$  for air-sea CO<sub>2</sub> flux estimates or ocean acidification studies (Figure 14).

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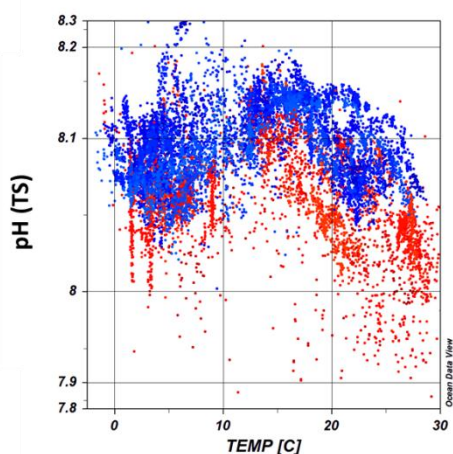
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966 **Figure 14:** An example of observed ocean acidification derived from the SNAPO-CO<sub>2</sub>-v2 dataset: pH (TS)  
967 calculated with  $A_T$  and  $C_T$  data are presented as a function of temperature (°C) for years 1998-2002 (blue  
968 symbols) and 2020-2023 (red symbols) and for salinity > 33 (Nb data selected with flag 2 = 11994). In recent  
969 years the pH was lower. Figure produced with ODV (Schlitzer, 2018).

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971 In almost all regions the new data in 2021-2023 indicated that the  $C_T$  concentrations were higher in  
972 recent years. In regions where data are available for more than 2 decades, the time-series show an increase of sea  
973 surface  $C_T$  (North Atlantic, Southern Indian Ocean and Ligurian Sea) with a rate close to or higher than the  
974 changes expected from anthropogenic CO<sub>2</sub> uptake. It is also recognized that at seasonal scale the  $C_T$  trends could  
975 be different. However, with the data in hand, the long-term trend of  $C_T$  cannot be quantified with confidence to  
976 compare with the anthropogenic carbon uptake in some regions. This is the case in the eastern tropical Atlantic  
977 subject to high inter-annual variability (Lefèvre et al., 2021, 2024) although new data have been added over  
978 2005-2022 in this region (Table 1, Figure S9). When data are available for less than a decade the increase in  $C_T$   
979 was observed but the trend was uncertain due to large inter-annual variability (e.g. Adélie Land). An exception  
980 was identified in the coastal zone in the Gulf of Lion (Mediterranean Sea) where summer data since 2010 present

981 a decrease in  $C_T$  most pronounced since 2015 ( $C_T$  trend =  $-5.2 \pm 1.5 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ). Such  $C_T$  decrease over 10  
982 years was also observed at the Hawaii Ocean Time series, HOT over 2010-2020 (Dore et al, 2009,  
983 <https://hahana.soest.hawaii.edu/hot/hotco2/hotco2.html>, last access: 27 August 2024).

984 Although the  $A_T$  concentrations present significant inter-annual variability such as in the NASPG, in the  
985 Topical Atlantic or the Adélie land and coastal zones,  $A_T$  appears relatively constant over time except at these  
986 locations. In the open ocean, we observed an increase of  $A_T$  in the Southern Ocean south of the Polar Front  
987 around  $60^\circ\text{S}$  in 2003-2012 not directly linked to salinity. In the coastal zones a decrease of  $A_T$  was pronounced  
988 south of Greenland. In the coast in the Gulf of Lion, as observed for  $C_T$ ,  $A_T$  decreased ( $A_T$  trend =  $-2.8 \pm 1.2 \mu\text{mol}$   
989  $\text{kg}^{-1} \text{yr}^{-1}$ ). This is opposed to the changes observed in the Ligurian Sea at station SOMLIT-Point-B, where  $C_T$  and  
990  $A_T$  increased over 2007-2015 (Kapsenberg et al, 2017) highlighting the contrasting  $C_T$  and  $A_T$  trends in the  
991 Mediterranean coastal zones where ocean acidification is detected (here over 2008-2022, pH trend of  $-0.048$   
992  $\pm 0.003 \text{decade}^{-1}$ ). With the continuous warming, reduced stratification and the rapid pH change observed in the  
993 Mediterranean Sea, how the marine ecosystems will respond in the future should be addressed (e.g. Howes et al,  
994 2015; Maugendre et al 2015; Lacoue-Labarthe et al, 2016). The SNAPO-CO2-v2 dataset could also be used to  
995 explore and analyze the changes of the carbonate system occurring during extreme events such as marine heat  
996 waves, rapid freshening, deep convection or high phytoplankton bloom events.

997 This dataset could also serve for validating autonomous platforms capable of measuring pH and  $f\text{CO}_2$   
998 properties (Sarmiento et al, 2023) and, along with other synthesis products (Jiang et al, 2024 in prep.), provides  
999 an additional reference dataset for the development and validation of regional biogeochemical models for  
1000 simulating air-sea  $\text{CO}_2$  fluxes. Thanks to the RECCAP2 stories, it has been recognized that Ocean  
1001 Biogeochemical Models present biases in the seasonal cycle of  $C_T$  and  $A_T$  due to inadequate representation of  
1002 biogeochemical cycles (e.g. Hauck et al, 2023; Rodgers et al, 2023; Sarma et al, 2023; Pérez et al, 2024;  
1003 Resplandy et al, 2024). The SNAPO-CO2-v2 dataset could be used to guide analyses for regional or global  
1004 biogeochemical models for  $A_T$  and  $C_T$  comparison and validation from seasonal to decadal scales. Our dataset is  
1005 also essential for training and validating neural networks capable of predicting variables in the carbonate system  
1006 (e.g. Fourier et al, 2020; Chau et al, 2024a; Gregor et al, 2024), thereby enhancing observations of marine  $\text{CO}_2$  at  
1007 different spatial and temporal scales. Furthermore, we encourage the use of this dataset (or part of it), at sea or  
1008 prior going to sea for cruise planning. Indeed, using the approach of Davis and Goyet (2021) which takes into  
1009 account the multiple constraints (ship-time, number of samples, etc.), it is possible to determine the most  
1010 appropriate sampling strategy (Guglielmi et al., 2022, 2023), to reach the specific scientific objectives of each  
1011 cruise.

1012 The data presented here are available online on the Seano server (<https://doi.org/10.17882/102337>) in a  
1013 file identifying version v1 and v2. The sources of the original datasets (doi) with the associated references are  
1014 listed in the Supplementary Material (Tables S3, S4). As for version v1 we invite the users to comment on any  
1015 anomaly that would have not been detected or to suggest potential misqualification of data in the present product  
1016 (e.g. data probably good although assigned with flag 3, probably wrong). As for SOCAT or GLODAP, we  
1017 expect to update the SNAPO-CO2 dataset once new observations are obtained and controlled.

1018  
1019 **7 Data availability**

1020 Data presented in this study are available at Seanoe (Metzl et al, 2024d, <https://www.seanoe.org>,  
1021 <https://doi.org/10.17882/102337>. See also <https://doi.org/10.17882/95414> for version V1. The dataset is also  
1022 available at <https://explore.webodv.awi.de/ocean/carbon/snapo-co2/>

1023  
1024 *Author contributions.* NM prepared the data synthesis, the figures and wrote the draft of the manuscript with  
1025 contributions from all authors. JF measured the discrete samples since 2014, with the help from CM and CLM,  
1026 and prepared the individual reports for each project. NM and JF pre-qualified the discrete  $A_T/C_T$  data. CLM and  
1027 NM are co-Is of the ongoing OISO project and qualified the underway  $A_T/C_T$  data from OISO cruises. FT and  
1028 CG were PIs of the MINERVE cruises. All authors have contributed either to organizing cruises, sample  
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1068

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