1	Temporal evolution of anthropogenic carbon in the subpolar
2	North Atlantic gyre between 2011 - 2021
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11 Key Points:

12	•	Anthropogenic carbon (C_{ant}) is studied with monthly time-series built from Argo-O ₂ data,
13		neural networks and a back calculation method.
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15	•	The Cant uptake in the first 2000 dbar of the Labrador and Irminger Seas increased by
16		$1.63\pm0.32\%$ yr ⁻¹ and $1.49\pm0.30\%$ yr ⁻¹ , respectively.
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18	٠	Over our study period, the long-term C _{ant} increase is modulated by ocean dynamics,
19		especially by deep winter convection.
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25 Abstract

The ocean plays a major role in the moderation of anthropogenically-induced climate change by 26 absorbing roughly a quarter of anthropogenic CO₂ (C_{ant}). This absorption of C_{ant} by the ocean 27 leads to ocean acidification, threatening marine's life. The North Atlantic Ocean encompasses 28 the highest ocean storage capacity of Cant per unit area. The subpolar North Atlantic gyre is 29 subject to a large seasonal to decadal variability that might impact Cant storage. To investigate 30 Cant evolution over the 2011-2021 period and its relationship with ocean dynamics in this region, 31 we use the Argo-O₂ array combined with neural networks and a back-calculation method (ϕC_T^{O} 32 method). We compute monthly time-series of Cant in the Labrador and Irminger Seas. We show 33 that Cant concentrations in the first 2000 dbar of the Labrador and Irminger Seas are strongly 34 affected by winter deep convection, especially between winter 2015 and winter 2018. The C_{ant} 35 inventories in the top 2000 dbar of the Labrador and Irminger Seas increase through time, at 36 rates of 1.63±0.32% yr⁻¹ and 1.49±0.30% yr⁻¹, respectively. Our monthly Argo-based C_{ant} 37 estimates complement high-quality ship-based measurements acquired at a biennial or lower 38 39 frequency. Additionally, this study shows that C_{ant} concentrations and C_{ant} inventories in deep convection areas may depend on the method employed to calculate Cant. As a consequence, we 40 take over the model ensemble idea and propose to use several methods to compute Cant, which 41 would give its methodological uncertainty. 42

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44 Plain Language Summary

45 Human activities have emitted large amounts of carbon dioxide (CO_2) into the atmosphere since the beginning of the industrial revolution. The global ocean has absorbed roughly a quarter of 46 these emissions, acting therefore as a moderator of global warming. The North Atlantic Ocean is 47 a region where most of this human-emitted CO_2 (C_{ant}) is uptaken by the ocean and stored. To 48 date, in the North Atlantic Ocean, oceanic Cant has only been studied via the coordination of 49 scientific cruises. These cruises collect water samples and perform accurate estimates of C_{ant} but 50 they have a short duration and are infrequent. To overcome this issue, we use autonomous Argo 51 floats, which are robotic profiling floats capable of measuring temperature, salinity and dissolved 52 oxygen every 10 days. We focused on the top 2000 meters of the Labrador and Irminger Seas 53 54 and show that their Cant contents are similar and increase over time. Furthermore, we show that

the vertical C_{ant} distribution is highly affected by ocean dynamics and, in particular, by intense winter mixing. Finally, our results demonstrate that C_{ant} values may vary depending on the method employed to compute it. As a consequence, we propose to calculate C_{ant} with several methods.

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60 **1 Introduction**

Since the beginning of the industrial revolution, human activities have emitted large amounts of 61 carbon dioxide (CO_2) in the atmosphere via fossil fuel burning, cement production, deforestation 62 and changes in land-use. This excess of atmospheric CO₂ is commonly distinct as anthropogenic 63 carbon (C_{ant}). The effective radiative forcing generated by this addition of C_{ant} in the atmosphere 64 corresponds to 2.28 W m⁻² over the 1979-2023 period (Forster et al., 2024). Via air-sea gas 65 exchange, the ocean has taken up to 26±5% of the total Cant emissions (Friedlingstein et al., 66 2023), acting therefore as a moderator of anthropogenically-induced climate change. Over the 67 1985-2018 period, a model ensemble study indicated that this uptake represents an addition of 68 2.1-2.4 PgC yr⁻¹ of C_{ant} in the ocean (DeVries et al., 2023). Even though C_{ant} represents less than 69 3% of the total dissolved inorganic carbon (DIC) pool (Guallart et al., 2015), the uptake and 70 storing of C_{ant} come at a cost for the ocean's ecosystem. With a vast impact on the chemical 71 oceanic properties, the major ecological footprint of Cant is ocean acidification, which lowers 72 oceanic pH and reduces the capacity of calcifying organisms to build and maintain their shells 73 (Doney et al., 2020). It is therefore primordial to better document the progression of Cant in the 74 ocean to establish where ocean acidification will have the most significant impact on the marine 75 biota and coral reefs. 76

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Two approaches are often used to compute oceanic C_{ant} . The first one gathers techniques collectively called back-calculation methods, that are based on the premise that C_{ant} concentration can be isolated from measured DIC. This is done by subtracting the DIC changes due to biological activity and by removing an estimate of the preindustrial preformed DIC concentration from the measured DIC. The second approach is the transit time distribution (TTD) method (Hall et al., 2002; Waugh et al., 2006). It relies on the assumption that C_{ant} penetrates the ocean as a passive tracer, responding to an evolving history in surface waters. Most importantly, the TTD method assumes an ocean circulation in steady state. While both methods provide similar spatial patterns, the TTD method tends to provide higher C_{ant} values than the back-calculation methods in deep convection areas (Vazquez-Rodriguez et al., 2009). All these methods have been widely applied to ship-based measurements (e.g., Gruber et al., 2019; Khatiwala et al., 2013; Sabine et al., 2004) to reconstruct C_{ant} climatologies (e.g., GLODAPv2; Lauvset et al., 2016) or to compute the global C_{ant} content over the oceans (Khatiwala et al., 2013; Sabine et al., 2004).

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The North Atlantic Ocean is an area of high Cant uptake and storage relative to its size (Davila et 93 al., 2022; Gruber et al., 2019; Pérez et al., 2024), enclosing 23-38% of the total oceanic Cant 94 content (Sabine et al., 2004; Steinfeldt et al., 2009). This is due to the combination of (1) a large 95 amount of Cant advected by the North Atlantic Current (NAC) from the subtropics (Brown, 96 97 McDonagh, Sanders, Watson, Wanninkhof, King, Smeed, Baringer, Meinen, Schuster, & others, 2021; Pérez et al., 2013) and (2) downward mixing that causes subduction of high C_{ant} surface 98 99 signal towards the ocean interior (Asselot et al., 2024; Pérez et al., 2018; Sabine et al., 2004). Mirroring the ongoing increase in atmospheric pCO_2 , the water-column C_{ant} content (C_{ant} 100 inventory) is increasing in the North Atlantic Ocean due to oceanic uptake (Pérez et al., 2008, 101 2010). In particular, the Labrador and Irminger Seas are considered as regions where Cant 102 103 accumulates through time. For instance, in the central Labrador Sea, Cant inventory has increased by 67% (0.6 PgC) between 1986 and 2016 (Raimondi et al., 2021). In the Irminger Sea, the 104 averaged C_{ant} concentration in the deep layers (potential density $\sigma_0 > 27.88$ kg m⁻³) has increased 105 by 76% (9.7 µmol kg⁻¹) between 1981 and 2006 (Pérez et al., 2010). Such estimates were made 106 possible owing to ship-based measurements and, in particular for the Irminger Sea, to the 107 biennial repetition of the GO-SHIP A25 OVIDE programme cruise. OVIDE has provided a 108 unique Cant time-series in the Irminger Basin for two-decades (García-Ibáñez et al., 2016; Pérez 109 et al., 2008, 2018). Such time-series highlighted the relationship between the deep-convection 110 periods that prevailed in the Irminger Sea over 2012-2018 (Zunino, et al., 2020) and the deeper 111 penetration of C_{ant} in this basin, compared to the shallower convection period in the early 2000s 112 (Pérez et al., 2018). 113

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115 Previously, Asselot et al. (2024) demonstrated the possibility to infer C_{ant} concentration by

combining Argo-O₂ data with neural networks and the back-calculation $\phi C_T^{\ O}$ method (Pérez et 116 al., 2008; Vazquez-Rodriguez et al., 2009). While less accurate than ship-based estimates, the 117 high temporal resolution of Argo-based Cant estimates and their spatial coverage over the entire 118 basins are an opportunity to extend the Cant estimates in between ship-based measurements. In 119 addition, Argo-based Cant estimates rely on neural networks that predict biogeochemical 120 variables based on water mass characteristics. Hence, Argo-based Cant estimates are able to 121 capture changes in C_{ant} in regions where water masses move laterally due to mesoscale processes 122 or rapid circulation changes. Taking advantage of the full spatio-temporal coverage of the Argo-123 O2 array and the efforts conducted since the early 2010s to maintain the network in the subpolar 124 North Atlantic gyre (Figure 1), the purpose of this study is to refine the understanding of C_{ant} 125 distribution at the finest spatio-temporal scale to date. This study focuses on the Irminger and 126 Labrador Seas and provides a monthly time-series of C_{ant} estimates over the period 2011-2021. 127 These unique time-series are used to describe the temporal evolution of C_{ant} concentrations and 128 inventories in the two basins and to investigate the relationship between the ocean dynamics and 129 Cant evolution at seasonal to interannual time-scales. 130

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132 **2 Data and methods**

133 $2.1 \operatorname{Argo-O_2} floats$

134 Our analysis is based on pressure (P), temperature (T), salinity (S) and oxygen (O_2) data from Argo-O₂ floats downloaded in December 2021 from the Argo Global Data Assembly Center 135 (Argo, 2000). To delimit the Irminger and Labrador Seas we consider the area of the basins 136 where depth exceeds 1700 dbar. Consequently, we selected only Argo-O₂ profiles within the 137 isobath 1700 dbar, resulting in 1387 and 1176 Argo-O₂ profiles in the Labrador and Irminger 138 Seas, respectively, spanning from January 2011 to December 2021 (Figure 1). We only used data 139 adjusted in delayed mode with a quality flag of 1 or 2 ("good" or "probably good" data) (Thierry 140 et al., 2021). The accuracy of the data is 0.02°C, 0.01, 2.4 dbar for temperature, salinity and 141 pressure, respectively (Wong et al., 2022), and better than 3 µmol kg⁻¹ for oxygen (Racapé et al., 142 2019). 143



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Figure 1. Location of the $Argo-O_2$ profiles and their distribution through time. The coloured circles indicate the temporal distribution of the Argo floats used in this study. The black stars in the Irminger Sea represent the location of the GO-SHIP A25 OVIDE stations. The black squares in the Labrador Sea indicate the location of the AR7W stations.

- 150
- 151 2.2 Bayesian neural networks
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As in Asselot et al. (2024), we rely on predictive neural networks to derive, from Argo-O₂ data, the biogeochemical and carbonate variables needed to calculate C_{ant} . Neural networks are machine learning algorithms based on a multi-layer perceptron composed of neurons which are elementary transfer functions (Bishop, 1995; Rumelhart et al., 1986). The neurons of a particular layer are connected with the neurons of the preceding and following layers by weights and biases that are readjusted during the training phases. The Bayesian approach introduces probability

distributions, meaning that a local uncertainty is given for each variable estimate. For this study, 159 two different neural networks are used. We apply ESPER_NN (Carter et al., 2021), which 160 considers T, S, O₂, geographical location and time to determine macronutrients (phosphate, 161 nitrate and silicate). This neural network is trained on the GLODAPv2.2020 (Olsen et al., 2020) 162 dataset plus a few additions at Gulf of Mexico and Mediterranean Sea. For the macronutrients, 163 ESPER_NN reproduces correctly the validation data, with averaged biases and errors of ~2%. 164 However, in the North Atlantic Ocean, ESPER_NN gives uncertainties of ~1.3% for carbonate 165 variables such as total alkalinity (A_T) and DIC, which is higher than previous neural networks. 166 As a consequence, we rely on CANYON-B and its associated routine CONTENT (Bittig et al., 167 2018) to estimate A_T and DIC. This particular routine ensures that the carbonate variables are 168 internally consistent within the carbonate system, giving A_T and DIC uncertainties of ~0.5%. 169 CANYON-B and CONTENT are trained and validated against GLODAPv2 data (Olsen et al., 170 2016) where 80% of these data are used for the training phase and the remaining 20% are used 171 for the validation procedure. 172

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2.3 Anthropogenic CO₂ estimates

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To isolate C_{ant} from the total oceanic carbon pool, we use the carbon-based back-calculation 176 method φC_T^{O} (Pérez et al., 2008; Vazquez-Rodriguez et al., 2009). This method is an upgraded 177 version of the ΔC^* method (Gruber et al., 1996), where the preformed A_T and air-sea 178 disequilibrium terms are obtained by using the subsurface layer (100-200 m) as a reference layer 179 to parameterize them. This assumption improves C_{ant} estimates in cold and deep water formation 180 regions subject to intense mixing such as the subpolar North Atlantic gyre. The ϕC_T^{O} method has 181 182 been broadly used to study Cant inventory, its storage rate (e.g. Pérez et al., 2008, 2010) and 3D spatial variability in the subpolar North Atlantic gyre (Asselot et al., 2024). The input variables 183 for this method are date, geographical location, pressure, T, S, O₂ (in this study, from Argo-O₂ 184 data), the macronutrients (in this study, from ESPER_NN), A_T and DIC (in this study, from 185 CONTENT). To quantify the uncertainty associated with our Cant concentrations, we randomly 186 generated 100 Cant fields using a Monte Carlo method (Metropolis & Ulam, 1949). The overall 187 averaged standard deviation from these C_{ant} fields is $\pm 3.9 \ \mu mol \ kg^{-1}$. Given this uncertainty, any 188

temporal changes in C_{ant} concentration lower than twice 3.9 µmol kg⁻¹ are not interpreted in our study.

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2.4 Estimation of the mixed layer depth

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194 To determine the mixed layer depth (MLD) in the Irminger and Labrador basins, we use the "threshold method" (de Boyer Montégut et al., 2004). This method is based on a density 195 difference between the ocean surface and the base of the mixed layer. For our study area, this 196 threshold value is set to 0.01 kg m⁻³ (Piron et al., 2016). To compute the monthly mean and 197 monthly maximum MLD, first, we binned the MLD values over a 2°x2° horizontal grid and 198 computed the average and maximum values included in the same grid cell and covering the same 199 month. As a result, we obtained a single average and maximum MLD value per month and per 200 grid cell. Second, we computed the mean and maximum MLD values per basin to obtain 201 monthly time series. 202

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2.5 Data analysis

To study the temporal evolution of C_{ant} during the 2011-2021 period, we looked at its 206 concentration over the first 2000 dbar and computed its inventory in the Irminger and Labrador 207 208 Seas. We vertically interpolated our Argo-O₂ data over 0-2000 dbar with regular spacing of 5 dbar. Then we gridded the Argo- O_2 profiles over a 2°x2° horizontal grid by calculating the 209 average of all the profiles included in the same grid cell and covering the same month. As a 210 result, we obtained an average C_{ant} profile per month and per grid cell. Finally, we calculated the 211 212 Cant inventory following the method of Tanhua & Keeling (2012) and Raimondi et al. (2021), given by Eq. 1: 213

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$$IC_{ant}(X,t) = \int_0^h C_{ant}(X,h,t) \cdot \rho \cdot dh$$

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where $IC_{ant}(X,t)$ stands for the water column inventory of C_{ant} (mol m⁻²) at location X and time t, $C_{ant}(X,h,t)$ is the concentration of C_{ant} (µmol kg⁻¹) at location X, depth h and time t, ρ is the *in-situ* density (kg m⁻³) and *dh* is the vertical spacing (m). To obtain the basin average C_{ant} inventories,

(1)

we selected all Argo- O_2 profiles in this basin for a particular month, calculated the monthly C_{ant} 220 inventory for each profile and averaged these values. As a result, we obtained a basin average 221 C_{ant} inventory for each month over the 2011-2021 period. The C_{ant} inventory is only computed in 222 the first 2000 dbar of the water column because most of our Argo-O₂ profiles do not descend 223 below this limit. The top-to-bottom Cant inventory was also computed in the Irminger Sea over 224 the period 2015-2021, owing to the availability of oxygen data from Deep-Argo float over the 225 full water column. To quantify the uncertainty associated with our Cant inventories, we randomly 226 generated 3000 Cant inventory profiles using a Monte Carlo method (Metropolis & Ulam, 1949). 227 The overall averaged standard deviation from these C_{ant} inventories is ±4.5 mol m⁻². From the 228 C_{ant} inventories, we calculated the storage rate (SR in mol m⁻² yr⁻¹) as a linear fit over the full 229 time series. The errors associated with the SR represent the root mean square error. 230

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In the Irminger Sea, C_{ant} inventory was also estimated from data acquired during the GO-SHIP A25 OVIDE cruises that collected full-depth hydrographic and biogeochemical measurements between Portugal and Greenland every two years since 2002. The C_{ant} concentrations from OVIDE data are computed with the ϕC_T^{O} method from A_T and pH, directly measured from bottle water samples (Zunino et al., 2014).

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2.6 Ocean	gridded C	Cant climato	ology
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We compare our Cant estimates with the existing GLODAPv2 gridded climatology (Lauvset et 240 al., 2016). This climatology is based on the GLODAP version 2 data (Olsen et al., 2016) product 241 covering all ocean basins over the years 1972 to 2013. To generate this climatology, the Data-242 243 Interpolating Variational Analysis (DIVA) was used, which allows a better processing of the topography compared to other interpolation methods. The climatological Cant field is computed 244 with a classical application of the TTD method (Hall et al., 2002; Waugh et al., 2006) on all 245 available CFC-12 data in GLODAPv2 product. This climatology is normalized to the year 2002 246 following the "atmospheric perturbation" concept (Ríos et al., 2012). 247

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To compare our data with the GLODAPv2 climatology (Lauvset et al., 2016), we need to scale this climatology to our study period. To scale these data, we use the method of Lauvset et al.

(2016). This method is based on the "atmospheric perturbation" concept (Ríos et al., 2012) that 251 assumes a transient steady state (Tanhua et al., 2007) where the accumulation of oceanic Cant is 252 related to the growth of C_{ant} in the atmosphere, as long as the atmospheric perturbation 253 progresses at an exponential rate. Thus, with knowledge on the atmospheric Cant perturbation 254 through time, the scaling method (Eq. 2) allows us to compute the oceanic C_{ant} at a certain time 255 from knowledge of Cant at a reference year. We note that the "atmospheric perturbation" concept 256 does not consider that the increase in oceanic DIC as atmospheric pCO_2 rises, diminishes over 257 time (i.e., Revelle effect ; Revelle & Suess, 1957). Consequently, this concept remains subject to 258 debate. Nevertheless, we perform our scaling over a narrow year interval, therefore we judge that 259 this assumption does not affect our results. 260

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$$C_{ant}^{T} = C_{ant}^{ref} + C_{ant}^{ref} \times \left(\frac{pCO_2^{T} - pCO_2^{ref}}{pCO_2^{ref} - pCO_2^{PI}}\right)$$
(2)

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where C_{ant}^{T} is the local oceanic C_{ant} concentration (µmol kg⁻¹) at time *T*, C_{ant}^{ref} is the local oceanic C_{ant} concentration (µmol kg⁻¹) at the reference year, pCO_2^{T} and pCO_2^{ref} are the atmospheric pCO_2 concentrations (µatm) at time *T* and at the reference year, respectively and, pCO_2^{PI} is the atmospheric pCO_2 concentration at pre-industrial times (i.e. 280 µatm).

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- 2.7 Method validation
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To validate our Cant estimates, we compare our Argo-O2-based Cant data with the GLODAPv2 271 climatology (Lauvset et al., 2016). For this comparison, we selected a particular oceanic layer, 272 representing the Labrador Sea Water (LSW) and delimited by $27.71 < \sigma_{\theta} < 27.80$ kg m⁻³ (Figure 273 2). This layer is located between 400-2000 dbar in the Labrador and Irminger Seas, and between 274 1000-1800 dbar in the rest of the subpolar gyre (Yashayaev & Loder, 2016). Regardless of the 275 vear considered, both datasets illustrate similar spatial patterns in the subpolar North Atlantic 276 gyre. They show a gradual C_{ant} increase from the southeast of the subpolar gyre to the northwest 277 of the gyre. Additionally, Cant concentrations is higher on the western side of the Reykjanes 278 Ridge compared to its eastern side. The mean Cant concentration is higher in the Labrador Sea 279 compared to the Irminger Sea. For instance, in 2013, the mean Cant concentration calculated 280

from Argo-O₂ in the Labrador Sea was $36.2\pm3.9 \text{ }\mu\text{mol} \text{ }kg^{-1}$ and $34.9\pm3.9 \text{ }\mu\text{mol} \text{ }kg^{-1}$ in the 281 Irminger Sea (Figure 2a). For the same year, the GLODAPv2 dataset (Lauvset et al., 2016) 282 indicates a mean C_{ant} concentration of 45.6 µmol kg⁻¹ in the Labrador Sea and 43.4 µmol kg⁻¹ in 283 the Irminger Sea (Figure 2c). The main difference between the two datasets is the magnitude of 284 the C_{ant} values, with higher concentrations in the GLODAPv2 climatology (Lauvset et al., 2016) 285 than in the Argo-based estimates. This difference is likely due to the different methods used to 286 compute C_{ant} . We used the ϕC_T^{O} method while Lauvset et al. (2016) used the TTD method. The 287 latter is known to overestimate Cant concentrations in deep water mass formation regions, such as 288 the subpolar North Atlantic gyre (Vazquez-Rodriguez et al., 2009). Previously, Asselot et al. 289 (2024) showed that C_{ant} estimates from GLODAPv2 are larger than the C_{ant} estimates based on 290 OVIDE bottle data, while the Argo-based and ship-based estimates are in good agreement when 291 calculated with the ϕC_T^{O} method. In addition, this difference in C_{ant} magnitude between both 292 datasets could come from the scaling method (Eq. 2) used on the GLODAPv2 climatology 293 (Lauvset et al., 2016). This method is based on the assumption of a steady increase of oceanic 294 C_{ant}, although this assumption remains subject to debate. 295



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subpolar North Atlantic gyre (35-66°N; 5-65°W). Argo-O₂ data (a) spanning the period 2011-2015 but normalized to the nominal year 2013 via Eq. 2 and (b) spanning the period 2015-2021 but normalized to the nominal year 2018 via the same equation. The Argo-O₂ data are gridded over a $2^{\circ}x2^{\circ}$ grid and the grid cells containing less than 10 Argo-O₂ profiles are coloured in gray. GLODAPv2 data (Lauvset et al., 2016) where C_{ant} is calculated with the TTD method and normalized to the year (c) 2013 and (d) 2018 via Eq. 2.

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306 **3 Results**

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3.1 Convective activity in the Irminger and Labrador basins

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310 To analyze the convective activity between January 2011 and December 2021, we rely on the mixed layer depth (MLD). In both basins, the MLD follows a seasonal signal. In summer, the 311 MLD is close to the ocean surface, then it deepens in autumn to reach a maximum depth in 312 winter and, finally, shallows in spring. However, the amplitude of this seasonal signal varies 313 314 through time (Figure 3). In the Irminger Sea, between 2011-2021, a maximum MLD of 2060 m was reached in April 2017 (Figure 3a). Between 2014 and 2018, the mean winter MLD always 315 316 exceeded 1300 m and the maximum winter MLD always exceeded 1700 m. This result indicates winters with intense convective activities during the 2014-2018 period in this basin. However, 317 318 between 2019 and 2021, the mean winter MLD reached ~500 m, suggesting a reduced convective activity during this period. In the Labrador Sea, a maximum MLD of 2030 m 319 320 occurred in March 2019 (Figure 3b). Throughout the whole study period, the maximum winter MLD always exceeded 1500 m except for winter 2013. However, the mean MLD illustrates two 321 322 distinct convective periods. The first one between winter 2014 and winter 2015 and the second between winter 2018 and winter 2021, corresponding to periods where the mean winter MLD 323 exceeded 1500 m. 324



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Figure 3. (a) Mixed layer depth (MLD) in the Irminger Sea. The dashed blue curve represents the maximum MLD while the green curve indicates the mean MLD. (b) MLD in the Labrador Sea. The dashed purple curve represents the maximum MLD while the orange curve indicates the mean MLD. Data are monthly-averaged. The Irminger and Labrador Seas are defined as the area of the basins where depth exceeds 1700 dbar.

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- 3.2 Monthly C_{ant} concentrations
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In the Irminger Sea and Labrador Sea, the ocean surface contains the highest Cant concentration 335 while these values decrease with depth (Figure 4a and 4b). For instance, in the first 500 dbar of 336 the Irminger Sea, the mean Cant concentration between January 2011 and December 2021 is 337 49.1 \pm 3.9 µmol kg⁻¹ while it is 33.9 \pm 3.9 µmol kg⁻¹ between 500-2000 dbar. For the Labrador Sea, 338 the mean C_{ant} concentration in the top 500 dbar over the whole study period is 47.0±3.9 µmol kg⁻ 339 ¹, decreasing to 37.9±3.9 µmol kg⁻¹ between 500-2000 dbar. Additionally, the mean C_{ant} 340 concentration increases over time in the two basins. For instance, in 2011, the yearly-mean 341 concentration in the top 100 dbar of the Irminger Sea reached $53.2\pm3.9 \mu$ mol kg⁻¹ and increased 342 to $61.4\pm3.9 \ \mu mol \ kg^{-1}$ in 2021. A linear regression through the study period indicates that this 343 increase represents an increase of 0.72±0.16 µmol kg⁻¹ yr⁻¹. In 2011 the yearly-mean Cant 344 concentration in the top 100 dbar of the Labrador Sea was 50.4±3.9 µmol kg⁻¹, increasing to 345 57.1 \pm 3.9 µmol kg⁻¹ in 2021. This increase represents an increase rate of 0.76 \pm 0.13 µmol kg⁻¹ yr⁻¹ 346

347 during the whole study period, which is slightly higher than the C_{ant} increase in the first 100 dbar of the Irminger Sea. At depth, although Cant concentrations are lower than at the surface, the rate 348 of Cant increase over time is slightly higher than at the surface for both basins. For instance, at 349 1400 dbar, the mean Cant concentration in the Irminger Sea was 32.0±3.9 µmol kg⁻¹ in 2011, 350 rising to 37.4±3.9 µmol kg⁻¹ in December 2021, representing an increase of 0.86±0.19 µmol kg⁻¹ 351 yr⁻¹. In the Labrador Sea, at 1400 dbar, the mean C_{ant} concentration was 35.0±3.9 µmol kg⁻¹ in 352 2011, increasing to 41.1 \pm 3.9 µmol kg⁻¹ in 2021. This increase represents an annual increase rate 353 of $0.74\pm0.15 \text{ }\mu\text{mol kg}^{-1} \text{ yr}^{-1}$ which is smaller than the annual increase rate at 1400 dbar in the 354 Irminger Sea. 355

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Between winter 2015 and winter 2018, a period of deep convection for the Irminger Sea (Nelson 357 et al., 2024; Zunino, et al., 2020), the isopycnal 27.71 kg m⁻³ shallowed and fluctuated around 358 ~250 dbar in this basin (Figure 4a). Prior to this period, the iso C_{ant} 40 µmol kg⁻¹ was above 1000 359 dbar, but during 2015-2018, it deepened to ~1500 dbar. After 2018, the isoCant 40 µmol kg⁻¹ 360 gradually returned to ~1000 dbar. Additionally, during the 2015-2018 period, the isoCant 45 µmol 361 kg⁻¹ followed a seasonal cycle, deepening to ~900 dbar in winter and rising to ~400 dbar during 362 summer. In the Labrador Sea, for winter 2015 and winter 2018, the isopycnal 27.71 kg m⁻³ 363 shallowed to levels above 100 dbar (Figure 4b). Simultaneously, the isoC_{ant} 40 µmol kg⁻¹ sharply 364 deepened, reaching ~1500 dbar in winter 2018. 365





Figure 4. Temporal evolution of C_{ant} concentration (µmol kg⁻¹) in the first 2000 dbar of the (a) 368 Irminger Sea and (b) Labrador Sea. The white lines depict the isopycnals $\sigma_{\theta} = 27.71$ kg m⁻³ and 369 $\sigma_{\theta} = 27.80 \text{ kg m}^{-3}$, delimiting the Labrador Sea Water. (c) Monthly evolution of C_{ant} inventories 370 (mol m⁻²) in the first 2000 dbar of the Irminger (light blue diamonds) and Labrador (light red 371 diamonds) Seas computed with Argo-O₂ data. The dark blue and dark red lines are obtained by 372 selecting the averaged Argo-based Cant inventories of 2011 and scaling it over the study period. 373 The scaling is computed via the "atmospheric perturbation" approach, representing the increase 374 in oceanic C_{ant} inventories if these inventories were only affected by the enhanced atmospheric 375 Cant concentrations (Eq. 2). 376



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During the study period, Cant inventories in the first 2000 dbar of the Labrador and Irminger Seas 380 increase through time (Figure 4c), indicating that both basins have accumulated and stored Cant 381 between 2011-2021. All along the study period, the Cant inventories in these two basins are close 382 to each other, indicating that both basins have a close Cant content. In the Irminger Sea, Argo-O₂ 383 data reveal that yearly-mean C_{ant} inventory in the top 2000 dbar was 75.9±4.5 mol m⁻² in 2011, 384 increasing to 84.2 ± 4.5 mol m⁻² in 2021. A linear regression through the whole study period 385 indicates that this increase represents a linear increase rate of 1.12 ± 0.22 mol m⁻² yr⁻¹. In the 386 Labrador Sea, Argo-O₂ data show that the top 2000 dbar contained 78.7 \pm 4.5 mol m⁻² of C_{ant} in 387 2011, increasing to 89.6 ± 4.5 mol m⁻² in December 2021. This increase outlines a linear increase 388 rate of 1.23 ± 0.24 mol m⁻² yr⁻¹, which is slightly higher than the increase in C_{ant} inventories for 389 the Irminger Sea. Between January 2011 and winter 2015, Cant inventories in the Labrador Sea 390 are on average higher by 2.16 mol m^{-2} than the ones in the Irminger Sea. In contrast, between 391 winter 2015 and winter 2018 which is an exceptional convective period in the Irminger Sea 392 (Figure 2), C_{ant} inventories are on average higher by 2.64 mol m⁻² in the Irminger Sea compared 393 to the ones in the Labrador Sea. After winter 2018, the Cant inventory of the Irminger Sea drops 394 back below the one of the Labrador Sea, with an averaged Cant inventories higher by 1.72 mol m 395 ² in the Labrador Sea compared to the Irminger Sea. We further compared the original Argo-396 based C_{ant} inventories with the scaled inventories (dark blue and dark red curve on Figure 4c). 397 These scaled data are derived by adjusting the average Argo-based Cant inventories from 2011 398 using the "atmospheric perturbation" approach (Ríos et al., 2012), which assumes that the 399 increase in oceanic Cant is solely due to the accumulation of Cant in the atmosphere (Eq. 2). In 400 other words, original Argo-based Cant inventories should follow the scaled data (Figure 4c) if 401 402 they were only related to the increased atmospheric Cant concentrations. However, the original Argo-based C_{ant} inventories show a lower increase than the scaled data. The increase rate of the 403 Argo-based Cant inventories are 1.12±0.22 mol m⁻² yr⁻¹ and 1.23±0.24 mol m⁻² yr⁻¹ in the 404 Irminger and Labrador basins, respectively while the scaled Cant inventories indicate an increase 405 rate of 1.59±0.02 mol m⁻² yr⁻¹ in both basins. This difference in increase rate between the 406 original and scaled inventories indicate that, originally, Argo-based Cant inventories are not only 407 affected by the atmospheric Cant perturbation. This result confirms the point that Cant inventories 408 are also affected by ocean dynamics and the buffer capacity of the ocean (Revelle factor). 409

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3.3.1 C_{ant} inventory in the Irminger Sea

- In the Irminger Sea, Argo-based Cant inventories are estimated either from the surface until 2000 413 dbar (light green points on Figure 5a) or from the surface until 3000 dbar (dark green points on 414 Figure 5b). In the first 2000 dbar, our results indicate that the C_{ant} inventory increased from 415 77 \pm 4.5 mol m⁻² in January 2011 to 83 \pm 4.5 mol m⁻² in December 2021, with a maximum 416 inventory of 90 ± 4.5 mol m⁻² in June 2018 (Figure 5a). Using a linear regression over the entire 417 study period, we estimated a storage rate of 1.12 ± 0.22 mol m⁻² yr⁻¹. However, during the 2016-418 2021 period, the storage rate of Cant in the top 2000 dbar of the Irminger Sea sharply decreased to 419 0.31 ± 0.18 mol m⁻² yr⁻¹. Deep-Argo data indicate that the C_{ant} inventory decreased from 112 ± 4.5 420 mol m⁻² in July 2016 to 103 ± 4.5 mol m⁻² in April 2021, with a maximum of 118 ± 4.5 mol m⁻² in 421 July 2018 (Figure 5b). For the Deep-Argo data, the storage rate is -0.35 ± 0.84 mol m⁻² yr⁻¹, 422 indicating that the first 3000 dbar of the water column loses Cant between July 2016 and April 423 2021. However, the Deep-Argo data have a short timespan and a small R-squared coefficient, 424 425 meaning that the value of the storage rate is highly debatable. Nevertheless, similar to the Deep-Argo data, the GO-SHIP A25 OVIDE data (brown line on Figure 5b) do not exhibit a particular 426 trend over the 2016-2021 period and show an averaged value of 110±4.5 mol m⁻², bringing 427 confidence in our Cant inventory values from Deep-Argo. Similar to previous studies (Fröb et al., 428 2016, 2018; Pérez et al., 2010), our Argo-based Cant inventories in the top 2000 dbar increase 429 over time (Figure 5a). The existing discrepancies in the magnitude of the inventories among 430 studies (Figure 5) are likely due to two factors. First, the area of the Irminger Sea considered 431 affects the C_{ant} inventories. For instance, Argo-O₂ data cover the whole Irminger basin (Figure 1) 432 433 while previous studies computed C_{ant} inventories along cruise sections only (Fröb et al., 2018; GO-SHIP A25 OVIDE) and have used these ship-based measurements to extrapolate over the 434 whole Irminger basin. Second, it depends on the method used to estimate Cant concentrations. As 435 in Fröb et al. (2018), we computed the C_{ant} concentrations with the φC_T^{O} method while Fröb et 436 al. (2016) employed the TTD method. 437
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dbar. The light green points represent the Argo-O₂ data calculated with the ϕC_T^{O} method (Pérez et al., 2008). The light blue curve represents the Cant inventory of Fröb et al. (2016) calculated with the TTD method (Waugh et al., 2006) over 100-2000 m. The orange curve represents the C_{ant} inventory from GO-SHIP A25 OVIDE data calculated with the ϕC_T^{O} method (Pérez et al., 2008). The pink diamond represents the GLODAPv2-based inventory calculated with the TTD method (Waugh et al., 2006). (b) Along the whole water column. The dark green points represent the Deep-Argo-O₂ data calculated with the φC_T^{O} method (Pérez et al., 2008). The dark blue curve indicates the C_{ant} inventory from Fröb et al. (2018) calculated with the ϕC_T^{O} method (Pérez 448 et al., 2008) from 100 m deep until the bottom of the water column for the part of the cruises' 449 transect between 40.5-31.5°W. The brown curve represents the Cant inventory from GO-SHIP 450 A25 OVIDE data calculated with the ϕC_T^0 method (Pérez et al., 2008). The pink diamond 451 represents the GLODAPv2-based inventory calculated with the TTD method (Waugh et al., 452 2006). SR = storage rate. The R-squared value represents the coefficient of determination of the 453 linear regression model. 454

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In the Labrador Sea, Argo-O₂ data show that the first 2000 dbar contained 81±4.5 mol m⁻² of C_{ant} 458 in July 2011, increasing to 90 ± 4.5 mol m⁻² in December 2021 (Figure 6). This increase outlines a 459 storage rate of 1.23 ± 0.24 mol m⁻² yr⁻¹, which is slightly higher than the storage rate in C_{ant} 460 inventories for the Irminger Sea. The maximum C_{ant} inventory reached 92±4.5 mol m⁻² in August 461 2021. Between July 2011 and February 2018, the Cant inventory follows a seasonal signal with 462 lower values in late autumn and higher values in spring/summer. On average this signal has an 463 amplitude of ~6 mol m⁻². However, Argo-O₂ data shows a sharp increase in C_{ant} inventory of 464 12.7% between March 2018 and July 2018. After this particular period, Cant inventory continues 465 to increase at a faster rate than before March 2018. The increase in Cant inventories depicted by 466 our Argo-O₂ data does not have the same trend, neither the same interannual variation and nor 467 the same mean value as the estimates of Raimondi et al. (2021). Several factors explain the 468 different magnitude in C_{ant} inventory between the Argo-O₂ data and the data of Raimondi et al. 469 (2021). First, our Argo-O₂ data cover the whole Labrador Sea (Figure 1) while Raimondi et al. 470 (2021) selected only the portion of the AR7W section where depth is superior to 3300 m. Thus 471 the results of Raimondi et al. (2021) cannot be extrapolated to the whole Labrador basin. Second, 472 with our Argo-O₂ data, C_{ant} is computed with the φC_T^O method while Raimondi et al. (2021) 473 employ the TTD method. However, the TTD method is known to overestimate Cant estimates in 474 water mass formation regions, such as the subpolar North Atlantic gyre (Vazquez-Rodriguez et 475 al., 2009), which may explain the higher values of Raimondi et al. (2021) compared to our Argo-476 477 O₂ estimates.



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Figure 6. Anthropogenic carbon inventory (mol m^{-2}) in the top 2000 dbar of the Labrador Sea. 479 The green diamonds represent the C_{ant} inventory from the Argo-O₂ data are calculated with the 480 ϕC_T^{O} method (Pérez et al., 2008). The red line represents the C_{ant} inventory calculated with the 481 refined version of the TTD method (Raimondi et al., 2021) along the portion of the AR7W line 482 483 with bottom depth >3300 m. The pink diamond represents the GLODAPv2-based inventory calculated with the TTD method (Waugh et al., 2006). SR = storage rate. The R-squared value 484 represents the coefficient of determination of the linear regression model. 485 486

487 4 Discussion

Both monthly time series of Cant in the Labrador and Irminger Seas (Figure 4a and 4b) show that 488 surface C_{ant} concentrations increase over time, reflecting the ongoing atmospheric C_{ant} increase 489 due to anthropogenic activities. In the first 500 dbar of the water column, the mean Cant 490 concentration over the whole study period is higher in the Irminger Sea than in the Labrador Sea. 491 This is due to the transport of subtropical waters, where C_{ant} is constrained within the surface, by 492 the NAC towards the Iceland basin. These waters cross the Reykjanes Ridge separating the 493 Iceland and Irminger basins, leading to a high Cant concentration in the surface of the Irminger 494 Sea (Asselot et al., 2024). These high C_{ant} concentrations at the surface of the Irminger Sea are 495 then transported towards the Labrador Sea via the Western Boundary Current (WBC). In the 496

497 Labrador Sea, C_{ant} is homogeneously redistributed along the water column, explaining the higher 498 C_{ant} concentration at depth in the Labrador Sea compared to the Irminger Sea.

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500 Between winter 2019 and winter 2021, our results show a shallower convection activity in the Irminger Sea than in the Labrador Sea (Figure 3), indicating a disconnection between the two 501 basins. However, this result could be due to the limited number of Argo profiles and the poor 502 sampling of the typical convection zone of the Irminger Sea during this period. Recently, 503 504 Yashayaev (2024) showed a decline in convection depth in the Labrador Sea starting in 2019 505 while our results do not support this finding. For instance, in 2021, Yashayaev (2024) reported a convection depth of ~800 m while our results indicate a mean MLD of 1879 m for March 2021. 506 Three hypotheses could explain these discrepancies. First, Yashayaev (2024) focused on the 507 Central Labrador Sea (CLS), defined as "the region where above average winter surface heat 508 509 losses are likely to result in deep convection", while this study considers the Labrador Sea as the area where depth exceeds 1700 dbar. Second, from 2019 onwards, the Argo-O₂ floats provided 510 limited sampling of the CLS area (Figure S1-S3). Third, the deep convection area is not 511 necessarily the CLS area. 512

513

Over the 2011-2021 period, the Cant storage rate in the first 2000 dbar of the Irminger Sea was 514 1.12 ± 0.22 mol m⁻² yr⁻¹ while it was 1.23 ± 0.24 mol m⁻² yr⁻¹ in the Labrador Sea, indicating that 515 both basins accumulate Cant over the last decade. Additionally, at the end of the study period, in 516 December 2021, the first 2000 dbar of the Irminger and Labrador Seas contained 83±4.5 mol m⁻² 517 and 90±4.5 mol m^{-2} of C_{ant}, respectively. Our Argo-O₂ data show that the Labrador Sea 518 contained more Cant and has a higher storage rate than the Irminger Sea. This might be due to the 519 deeper convective activity that happened in the Labrador Sea during our study period compared 520 521 to the convective activity of the Irminger Sea (Figure 3). Indeed, our Argo-O₂ data indicate that the mean MLD barely goes below 1500 dbar in the Irminger Sea while it exceeded this limit for 522 several winters in the Labrador Sea (Figure 3). In the Irminger Sea, the storage rate in the top 523 2000 dbar decreases between 2016 and 2021 (Figure 5a). We speculate that this reduction might 524 be due to a weakening of the Atlantic Meridional Overturning Circulation (AMOC) during this 525 526 period, cooling the eastern subpolar North Atlantic gyre (Bryden et al., 2020) and reducing the transport of Cant from the subtropics towards the subpolar gyre (Brown, McDonagh, Sanders, 527

Watson, Wanninkhof, King, Smeed, Baringer, Meinen, Schuster, Yool, et al., 2021). The higher 528 Cant inventories in the Irminger Sea compared to the Labrador Sea between winter 2015 and 529 winter 2018 (Figure 4c) can be explained by two hypotheses. First, the exceptional convective 530 activity during this period may have led to a C_{ant} redistribution deeper than 2000 dbar in the 531 Labrador Sea due to the deeper convective activity in this basin (Yashayaev, 2024; Zunino, et al., 532 2020) compared to the deep redistribution in the Irminger Sea. Second, this intense convective 533 period may have led to a larger Cant transport outside the Labrador Sea compared to the transport 534 outside the Irminger Sea. 535

536

In the Labrador Sea, the estimates of Raimondi et al. (2021) show that Cant inventory sharply 537 rose in July 2012 while our Argo-O₂ data does not support this result (Figure 6). However, with 538 the exception of 2013, our Argo-O₂ data indicate a maximum MLD reaching >2000 m in the 539 540 Labrador Sea between 2012 and 2016 (Figure 3). Additionally, previous studies also report above-average convective activity during these winters for this basin, with mixed layers reaching 541 ~1500 m (Kieke & Yashayaev, 2015; Yashayaev & Loder, 2016), supporting the rise in Cant 542 inventory between 2012 and 2016 reported by Raimondi et al. (2021). We argue that our Argo-543 O2 data do not depict an increase in Cant inventory between 2012 and 2016 due to marginal 544 sampling of the convection area (>2000 m; Figure S1 and S2), as indicated by the value of the 545 mean MLD (Figure 3). The marginal sampling could be attributed either to the small size of the 546 convection area or to poor sampling of the zone. However, in the Labrador Sea, the Argo-based 547 C_{ant} inventory sharply increases in winter 2018, which might be due to the exceptional 548 convective activity occurring at this period. Indeed, the maximum depth of convection in winter 549 2018 reached up to 1855 m (Figure 3), closely matching the 1866 m reported by Zunino et al. 550 (2020). This exceptional convective activity has, thus, redistributed C_{ant} along the water column 551 and has enhanced the air-sea Cant flux, explaining the sharp increase of Cant inventory in winter 552 and spring 2018 (Figure 6). In addition, our Cant inventories and Cant storage rate for the Labrador 553 Sea are lower than the ones of Raimondi et al. (2021). We assert that these differences between 554 our Argo-O₂ data and Raimondi et al. (2021) come from the method employed to calculate C_{ant} 555 and the area of the Labrador Sea considered. Indeed, Raimondi et al. (2021) only used data along 556 the portion of the AR7W line with bottom depth >3300 m (10 profiles per AR7W cruise or 210 557 profiles in total) while we use data covering the whole Labrador Sea (1387 profiles in total). 558

Moreover, Raimondi et al. (2021) employed the TTD method which is sensitive to assumptions 559 of saturation of water mass age tracers (e.g., CFCs). In contrast, we use a back-calculation 560 method that is based on oceanic DIC content and that does not require a water mass age tracer. 561 However, using the same method to compute C_{ant} with different databases provides comparable 562 Cant inventories. For instance, in the Irminger Sea, Cant inventories from Deep-Argo and GO-563 SHIP A25 OVIDE data (orange line on Figure 5), calculated with the ϕC_T^{O} method (Pérez et al., 564 2008), are in agreement. This result indicates that if Cant inventories are calculated with the same 565 method, our Argo-O₂ data are in agreement with high-quality ship-based measurements. 566 Consequently, our study highlights the fact that Cant values and trends may depend on the method 567 used to compute C_{ant}. 568

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570 The GLODAPv2 climatological product (Lauvset et al., 2016), gives a global overview of Cant distribution in the oceans. This product has been widely used to study e.g. the global pH 571 distribution (Lauvset et al., 2020), oceanic sink of Cant (Gruber et al., 2019; Ridge & McKinley, 572 2020) and to tune Earth system models (Terhaar et al., 2021). Moreover, this climatology permits 573 574 to study the regional Cant patterns in the North Atlantic Ocean (Figure 2). The GLODAPv2 data (Lauvset et al., 2016) illustrate the Cant gradient from the south-east of the subpolar North 575 Atlantic gyre to its north-west side, with the Reykjanes Ridge acting as a border between the 576 Iceland basin and the Irminger Sea. The Argo-O₂ data replicate this C_{ant} spatial distribution in the 577 North Atlantic Ocean. Using GLODAPv2 data (Lauvset et al., 2016) or Argo-O₂ data to study 578 the global or regional patterns of Cant will yield the same conclusions. However, to study the 579 temporal evolution of C_{ant}, Argo-O₂ data are more advantageous than the other data sources. For 580 instance, the GLODAPv2 data (Lauvset et al., 2016) are originally scaled to the nominal year 581 2002. To scale these data to another time period, we used the scaling equation of Lauvset et al. 582 (2016) who follow the "atmospheric perturbation" concept (Ríos et al., 2012), which is based on 583 the transient steady state (Tanhua et al., 2007). However, the transient steady state assumption 584 has been challenged and several studies demonstrate that the Cant uptake is not steady through 585 time (see Gruber et al. (2023) for a review). As a consequence, Lauvset et al. (2016) declare that 586 their scaling equation should not be applied to data records spanning more than 40 years due to a 587 potential decrease of the ocean sensitivity to atmospheric Cant perturbation through time. This 588

estimate of "40 years" might even be overestimated, as our findings reveal a divergence between
the original Argo data and the scaled Argo data after only 11 years (Figure 4c).

591

592 In a previous study, Gruber et al. (2019) determined that the C_{ant} inventory increased at a steady rate of 1.52% yr⁻¹ in the North Atlantic between 1994-2007. This steady C_{ant} increase assumption 593 has been primarily questioned by McNeil & Matear (2013). Using CO₂ observations and model 594 predictions, McNeil & Matear (2013) showed that the rate of oceanic CO₂ uptake is slowing, 595 596 largely due a natural decadal-scale outgassing signal. The authors even proposed a simple 597 concept to extract the non-steady state CO₂ signal from observations but their methodological uncertainty is too large at present to provide a significant non-steady state signal. Recently, 598 Müller et al. (2023) also challenged the steady C_{ant} increase approach by illustrating a reduction 599 of the global Cant uptake between 1994 and 2014. In the North Atlantic Ocean, Müller et al. 600 (2023) declare that this reduction in Cant increase is due to a lower buffer capacity, a more 601 stratified ocean and a weaker overturning circulation in the upper ocean. Even if our data do not 602 support the steady C_{ant} increase assumption (Figure 4c), by considering it, our Argo-O₂ data 603 indicate that the averaged Cant increase between 2011-2021 was 1.63±0.32% yr⁻¹ and 604 1.49±0.30% yr⁻¹ in the first 2000 dbar of the Labrador and Irminger Sea, respectively. To the 605 best of our knowledge, these values are the first ones for these two basins but they should be 606 607 treated with caution and used for data covering our study period only. These values are in close agreement with previous estimates. For instance, Steinfeldt et al. (2009) indicated that the Cant 608 increase was 1.69% yr⁻¹ between 1850-2003 in the latitudinal band 20°S-60°N of the Atlantic 609 Ocean. In addition, Gruber et al. (2019) suggested that the C_{ant} increase was 1.52% yr⁻¹ for the 610 North Atlantic Ocean between 1994-2007. More recently, Steinfeldt et al. (2024) specified that, 611 for the Atlantic Ocean, the C_{ant} increase was 1.74% yr⁻¹ for the decade 1990-2000, 1.73% yr⁻¹ for 612 2000-2010 and 1.68% yr⁻¹ for 2010-2020, illustrating a reduction in C_{ant} growth over time. The 613 different Cant increase values for the Labrador and Irminger Seas also indicate that Cant increase 614 varies in space, a point already emitted by Gruber et al. (2019). 615

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Most of the previous studies, investigating the distribution and evolution of C_{ant} , are based on repeat hydrographic cruises that limit the ability to constrain the yearly or monthly variability of the ocean C_{ant} sink (Gruber et al., 2023). For instance, ship-based measurements are most of the

time acquired at a bi-annual or lower frequency during summer months, neglecting any change 620 in C_{ant} during the winter months. The insufficient and uneven observations can even give strong 621 biases in the trend of the ocean carbon sink. For instance, it can give an overestimation of the 622 global CO₂ flux by 20-35% in pCO₂ products (Hauck et al., 2023). In contrast, Argo-O₂ data, 623 having a year-round coverage, enable to investigate Cant evolution during the whole year and 624 identify the winter processes affecting its distribution. For instance, with our Argo- O_2 data, we 625 are able to compute the monthly evolution of Cant inventory and link its evolution with the deep 626 convection period occurring in the subpolar North Atlantic gyre. This is even more relevant since 627 we know that intense winter convection activity contributes to the transport of C_{ant} towards the 628 deep layers of the ocean (Pérez et al., 2008). Argo-O2 data obviously increase the temporal 629 coverage of C_{ant} estimates (Figure 5 and 6) and help to understand the processes behind the C_{ant} 630 631 distribution. In addition, using Argo-O₂ data would consequently constrain C_{ant} yearly and monthly variability and thus resolve the non-steady-state variation in Cant. 632

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634 **5 Conclusions and global implications**

This study provides the first monthly reconstruction of Cant evolution in the Labrador and 635 Irminger Seas over the 2011-2021 period. So far, Argo-O₂ data must be combined with neural 636 networks to obtain Cant estimates, and thus these data crucially depend on bottle measurements 637 for the learning phase of neural networks. We were able to produce monthly time-series whereas, 638 previously, with ship-based measurements we only had a single data every year or two. In the 639 640 future, this unprecedented increase in spatiotemporal Cant coverage might help to study, identify and compare the different mechanisms affecting Cant evolution and distribution in the oceans on 641 a thin spatio-temporal scale. The higher spatiotemporal coverage of Argo may even allow for the 642 detection of interannual changes that cruise sections cannot capture. The results presented here 643 are limited to 2000 dbar because no sufficient Argo-O₂ floats sampled below this limit. The 644 OneArgo mission plans to deploy more than 20 Deep-Argo floats, capable of reaching 4000 or 645 6000 dbar, in the subpolar North Atlantic gyre (Roemmich et al., 2022). Using these Deep-Argo 646 floats could supplement our findings and unveil the Cant evolution below 2000 dbar. 647 648

649 Our methodological approach, linking $Argo-O_2$ floats, neural networks and a back-calculation 650 method, gives similar spatial C_{ant} patterns in the subpolar North Atlantic gyre than the

GLODAPv2 climatology (Lauvset et al., 2016). This similarity enables us to validate the 651 methodology presented here. However, this study shows that Cant concentrations and Cant 652 inventories may depend on the method employed to calculate Cant (e.g. back-calculation method 653 or TTD method), thus we cannot be satisfied to use only one method to estimate it. As a 654 consequence, we take over the model ensemble idea, widely accepted by the Earth system 655 model's community, and propose to use several methods to compute C_{ant} when we study this 656 biogeochemical variable. The average C_{ant} value obtained between the different methods might 657 be less influenced by the different pitfalls and shortcomings of each method. Additionally, this 658 view will provide the methodological C_{ant} uncertainty, which in the future needs to be reduced. 659

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674 **Open Research**

The original Argo data can be downloaded on the Euro Argo Data Selection platform. Our C_{ant} estimates, needed to evaluate the conclusion of the paper, can be downloaded on Zenodo (https://doi.org/10.5281/zenodo.7899993).

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