



The subsurface layer reference to calculate preformed alkalinity and air–sea CO₂ disequilibrium in the Atlantic Ocean

M. Vázquez-Rodríguez*, X.A. Padin, P.C. Pardo, A.F. Ríos, F.F. Pérez

Instituto de Investigaciones Marinas, CSIC, Eduardo Cabello 6, E-36208 Vigo, Spain

ARTICLE INFO

Article history:

Received 21 February 2011

Received in revised form 5 October 2011

Accepted 7 October 2011

Available online 20 October 2011

Keywords:

Preformed total alkalinity

Air–sea CO₂ disequilibrium

Carbon cycle

Anthropogenic CO₂

Atlantic Ocean

Subsurface layer

Parameterisation

ABSTRACT

The subsurface ocean layer (100–200 m deep) is suggested as a reference to parameterise preformed alkalinity (A_T°) and air–sea CO₂ disequilibrium (ΔC_{dis}) in the Atlantic. Results suggest that this domain retains the memory of water mass formation (WMF) conditions over annual periods and avoids the large, short-term variability of the uppermost layers. Its thermohaline variability also encompasses and represents all water masses that outcrop in the Atlantic. Subsurface data also avoids the scarcity of late wintertime surface observations and benefit from the larger availability of year-round measurements, thus enhancing their representativeness and application coverage. This last feature is most relevant in high Atlantic latitudes, where WMF typically occurs and the widespread ice sheets often preclude surface pCO₂ sampling during wintertime. The obtained A_T° and ΔC_{dis} parameterisations achieve uncertainties of ± 4.6 and $\pm 5.6 \mu\text{mol kg}^{-1}$, respectively, improving significantly the estimates in previous works, particularly in the high latitudes. The A_T° parameterisation is well correlated with observations and is coherent with the latitudinal subsurface distribution of silicate, particularly in the northern subpolar region, where previous studies showed discrepancies. The ΔC_{dis} estimates in the upper layers are coherent with air–sea $\Delta p\text{CO}_2$ data from Takahashi's climatology, thus tackling known important shortcomings and biases of anthropogenic CO₂ estimates in Atlantic waters.

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1. Introduction

In recent years, a good deal of coordinated international research programmes, like WOCE, JGOFS or CARBOOCEAN, has focused on tackling and minimising the uncertainties of the global carbon budget. These uncertainties stem from the difficulty of estimating accurately the air–sea CO₂ fluxes and carbon storage in some environmental compartments like the land and the ocean. A fundamental piece for solving the intricate puzzle of global carbon content is estimating the anthropogenic CO₂ (C_{ant}) that dissolves into the ocean.

The C_{ant} has built up and added to the natural fraction of the total inorganic carbon (C_T) pool in the ocean since the Industrial Revolution, when the zero-baseline for C_{ant} is set by definition. The molecules of C_{ant} and natural CO₂ are indistinguishable, and there is no analytical procedure at present that is able to quantify C_{ant} directly. Therefore, this has to be estimated indirectly from other measurable physical–chemical tracers, under certain assumptions. Back-calculation methods to estimate C_{ant} are based on the fact that preformed C_T (C_T° , the C_T stored in a given water mass when it is formed) has changed since the Industrial revolution due to the increasing partial pressures of atmospheric CO₂

($p\text{CO}_2^{\text{atm}}$), which forces more of this gas to dissolve in the ocean (Brewer, 1978).

The C_{ant} added to the naturally occurring fraction of C_T° in a water mass can be reckoned by subtracting from it (back-calculating) a pre-industrial “zero- C_{ant} ” reference: $C_{ant} = C_T^\circ - C_T^{\text{PI}}$ (superscript “PI” denotes “Pre-Industrial”). The estimates of back-calculated C_{ant} improved over the years from many contributions (Gruber et al., 1996; Pérez et al., 2002; Touratier et al., 2007; Wallace, 2001; Waugh et al., 2006). The first formal definition and estimation of the CO₂ air–sea disequilibrium term (ΔC_{dis}) by Gruber et al. (1996) meant a leap forward in the back-calculation technique, albeit with certain caveats (Matsumoto and Gruber, 2005). The C_{ant} was re-expressed as the difference between the quasi-conservative tracer ΔC^* and $\Delta C_{dis}^{\text{PI}}$, namely: $C_{ant} = \Delta C^* - \Delta C_{dis}^{\text{PI}} = (C_T^\circ - C_{T_{eq}}^{\text{PI}}) - \Delta C_{dis}^{\text{PI}}$, where $C_{T_{eq}}^{\text{PI}}$ is the C_T in equilibrium with the pre-industrial atmospheric CO₂ level of 280 ppm (calculated from thermodynamic equations) and $\Delta C_{dis}^{\text{PI}}$ is the ΔC_{dis} during the pre-industrial era.

When water masses upwell to the surface from lower layers the air–sea gas exchanges lead them to reach different degrees of equilibrium with the atmospheric partial pressure CO₂ ($p\text{CO}_2^{\text{atm}}$), depending on the existing air–sea pCO₂ gradient ($\Delta p\text{CO}_2$) during water mass ventilation. Afterwards, the water masses lose contact with the atmosphere as they sink (water mass formation; WMF) and air–sea exchanges cease. A certain amount of CO₂ gets stored in seawater, where it can remain in the inorganic phase or be incorporated through photosynthetic activity to the organic carbon pool. From this mechanism of water mass ventilation

* Corresponding author. Tel.: + 34 986231930; fax: + 34 986305305.

E-mail addresses: mvazquez@iim.csic.es (M. Vázquez-Rodríguez), padin@iim.csic.es (X.A. Padin), pconde@iim.csic.es (P.C. Pardo), aida@iim.csic.es (A.F. Ríos), fiz.perez@iim.csic.es (F.F. Pérez).

follows that one of the most crucial aspects for back-calculating C_{ant} from C_T -based techniques relies essentially in being able to reconstruct accurately WMF conditions from the available observations.

Unfortunately, cruises are usually unable to record WMF conditions as they do not normally coincide with that particular moment, and this leaves the representativeness of WMF conditions from the available datasets as a pending issue that limits the quality of parameterisations for preformed total alkalinity (A_T ; the A_T of a water mass when it outcrops and loses contact with the atmosphere) and ΔC_{dis} . The use of the available data should therefore be optimised in order to compensate for the spatiotemporal sparseness of observations and attain the best results possible. Recently, the work from Pardo et al. (2011) addressed this issue in the Pacific and Indian basins by proposing the use of subsurface data as a reference to parameterise A_T and ΔC_{dis} .

The complexity of the numerous deep-water mass formation processes hosted by the Atlantic Ocean represents an added difficulty for finding an analogous reference layer. The present study has a two-fold purpose: a) find and provide supporting evidence for a reference region in the Atlantic that retains the memory of WMF conditions and; b) use data from the Atlantic to obtain optimum estimates of A_T and ΔC_{dis} and compare them with previous estimates from different authors in order to evaluate the results.

2. Dataset

The current work uses a combination of three different datasets to study the Atlantic reference layer for A_T and ΔC_{dis} , namely: a) bottle

data from eleven different cruises; b) A subset of selected surface and subsurface data (potential temperature, salinity, nitrate and phosphate) from the World Ocean Atlas 2005 (WOA05) for three $5^\circ \times 5^\circ$ regions; c) Means of wintertime ΔC_{dis} calculated from $\Delta p\text{CO}_2$ values in the latest available $p\text{CO}_2$ climatology from Takahashi et al. (2009).

The selected cruises (Fig. 1; Table 1) include the WOCE tracks A02, A14, A16, A17, A20, AR01, I06-Sa and I06-Sb, the CLIVAR A16N legs 1 and 2, the WOCE/CLIVAR OVIDE 2002 and 2004 cruises and the NSeas-Knorr cruise (Bellerby et al., 2005; Olsen et al., 2006). The availability of high-quality, carbon system measurements, calibrated with Certified Reference Materials (CRMs) and quality-controlled were the main cruise selection criteria. All of the above cruises are part of the Atlantic Synthesis effort made within the CARBOOCEAN Integrated Project framework (<http://www.carbon-synthesis.org/>). The data are available from the Global Ocean Data Analysis Project (GLODAP; http://cdiac.ornl.gov/oceans/glodap/Glodap_home.htm), the Climate Variability and Predictability (CLIVAR; <http://www.clivar.org>) and the Carbon In the Atlantic (CARINA; <http://store.pangaea.de/Projects/CARBOOCEAN/carina/index.htm>) data portals.

For the vast majority of the samples, pressure and temperature data come from filtered CTD measurements. Salinity and nutrient data come from analysis of individual Niskin bottles collected with a rosette. All WOCE C_T samples in the dataset used were analysed with the coulometric titration technique described in Johnson et al., 1993. For the OVIDE cruise, C_T data was obtained from the thermodynamic equations of the carbon system using direct measurements of pH and A_T , and the carbon dioxide dissociation constants from Dickson and Millero (1987).

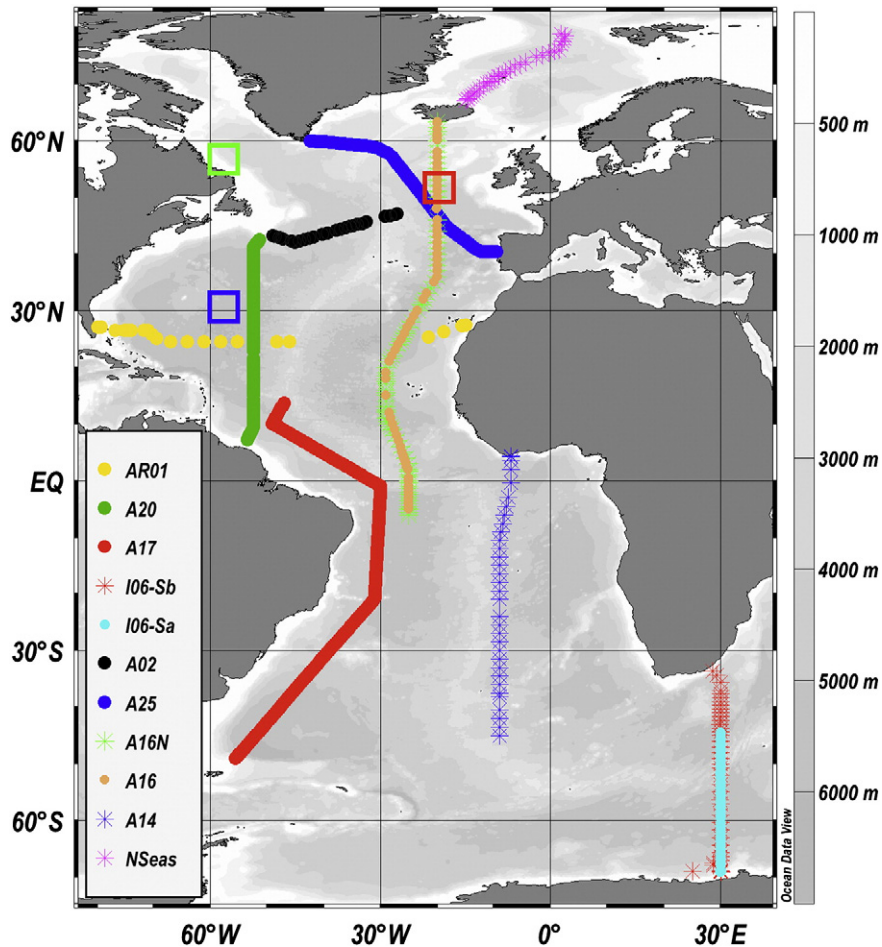


Fig. 1. Atlantic cruises used to test the adequacy of different reference layers for A_T and ΔC_{dis} parameterisations. Ancillary information for these cruises is given in Table 1. The Sargasso Seawater (SSW) formation area [(55.5° W–60.5° W);(28.5° N–33.5° N)], the Subpolar Mode Water (SPMW) formation area [(17.5° W, 22.5° W);(49.5° N, 54.5° N)] and the Labrador Seawater (LSW) formation area [(55.5° W, 60.5° W);(28.5° N, 33.5° N)], are framed by the blue, red and green squares, respectively. These regions are used as references to explore the annual variability of tracers in the surface and subsurface layers (Fig.2).

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