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Measurements of total alkalinity and inorganic dissolved carbon in the Atlantic Ocean and adjacent Southern Ocean between 2008 and 2010

U. Schuster[1,*], A. J. Watson[1,*], D. C. E. Bakker[2], A. M. de Boer[3,*], E. M. Jones[4,*], G. A. Lee[2], O. Legge[2], A. Louwerse[1,*], J. Riley[5], and S. Scally[*]

[1]College of Life and Environmental Sciences, University of Exeter, Exeter, EX4 4PS, UK
[2]Centre for Ocean and Atmospheric Science, School of Environmental Sciences, University of East Anglia, Norwich Research Park, Norwich, NR4 7TJ, UK
[3]Department of Geological Sciences and Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden
[4]Alfred Wegener Institute for Polar and Marine Research, Climate Sciences, Postfach 120161, 27515 Bremerhaven, Germany
[5]International CLIVAR Project Office, National Oceanography Centre, Southampton, Waterfront Campus, European Way, Southampton, SO14 3ZH, UK
[*]formerly at: School of Environmental Sciences, University of East Anglia, Norwich Research Park, Norwich, NR4 7TJ, UK

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Correspondence to: U. Schuster (u.schuster@exeter.ac.uk)

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Abstract

Water column dissolved inorganic carbon and total alkalinity were measured during five hydrographic sections in the Atlantic Ocean and Drake Passage. The work was funded through the Strategic Funding Initiative of the UK's Oceans2025 programme, which ran from 2007 to 2012. The aims of this programme were to establish the regional budgets of natural and anthropogenic carbon in the North Atlantic, the South Atlantic, and the Atlantic sector of the Southern Ocean, as well as the rates of change of these budgets. This paper describes the dissolved inorganic carbon and total alkalinity data collected along east-west sections at 55–60° N (Arctic Gateway), 24.5° N, and 24° S in the Atlantic and across two Drake Passage sections. Other hydrographic and biogeochemical parameters were measured during these sections, yet are not covered in this paper.

Over 95 % of samples taken during the 24.5° N, 24° S, and the Drake Passage sections were analysed onboard and subjected to a 1st level quality control addressing technical and analytical issues. Samples taken during Arctic Gateway were analysed and subjected to quality control back in the laboratory. Complete post-cruise 2nd level quality control was performed using cross-over analysis with historical data in the vicinity of measurements, and data are available through the Carbon Dioxide Information Analysis Center (CDIAC) and are included in the Global Ocean Data Analyses Project, version 2 (GLODAP 2).

Data coverage and parameter measured

Repository-References:
DI332 – doi:10.3334/CDIAC/OTG.CLIVAR_AR07W_74DI20080820
DI364 – doi:10.3334/CDIAC/OTG.CLIVAR_A05_2010
JC032 – doi:10.3334/CDIAC/OTG.CLIVAR_A9.5_2009
JC031 – doi:10.3334/CDIAC/OTG.CLIVAR_A21_JC031_2009

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Coverage: Atlantic Ocean 64.1° S to 60.6° N/79.9 to 11.1° W
In detail: DI332: 47.51 to 60.6° N/55.5 to 11.1° W
DI364: 23.3 to 27.9° N/79.9 to 13.4° W
JC032: 37.4 to 22.2° S/53.5° W to 13.7° E
JC031 (A21): 57.1 to 64.1° S/68.3 to 63.1° W, and (SR01b): 61.1 to 54.7° S/54.6 to 58.0° W, with an extension to 53.14° S 58.00° W
Dates: DI332: 20 August to 25 September 2008
DI364: 5 January to 19 February 2010
JC032: 7 March and 21 April 2009
JC031: 3 February and 3 March 2009

1 Introduction

The world oceans have taken up approximately half of the carbon dioxide (CO_2) released by the burning of fossil fuel since the Industrial Revolution. Without this sink of CO_2, the atmospheric content of CO_2 would be much higher than it is today. Moreover, the oceanic uptake of atmospheric CO_2 is not uniform in either time or space, so that the future concentration of atmospheric CO_2 is highly dependent on the behaviour and variability of this sink.

The control of the spatial and temporal variability of oceanic CO_2 uptake is not well understood. One of the major influences is the Meridional Overturning Circulation (MOC). The MOC, in particular the Atlantic MOC, therefore has a significant impact on climate via their influence on the carbon cycle.

In order to predict the future behaviour of the ocean sink, it is crucial to first get a better understanding of its present behaviour. To that end, sustained observations are needed of interior ocean biogeochemical and hydrographic parameters.

Between 2007 and 2012, a strategic research programme of the UK's National Environmental Research Council (NERC) Marine Centres, Oceans2025 (http://www.oceans2025.org/) was funded. *Theme 1: Climate, Ocean Circulation and Sea Level*,

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aimed to establish regional budgets of heat, freshwater, and carbon via the measurements of physical, chemical and biological parameters on a set of hydrographic sections in the Atlantic and the Atlantic sector of the Southern Ocean. To ensure high quality measurements of carbon and transient tracers on these sections, a Strategic Funding Initiative (SOFI) was funded titled "A carbon and transient tracer measurement programme in the Atlantic and Southern Ocean under Oceans2025". In this paper we describe the measurements of dissolved inorganic carbon (DIC) and total alkalinity (TA) performed on these sections.

These measurements were a UK-funded contribution to the international repeat hydrography effort (CLIVAR/GO-SHIP programmes, http://www.go-ship.org/).

2 Common sampling and analyses

As part of SOFI, DIC and TA were analysed in seawater samples on five hydrographic sections in the Atlantic Ocean and the Atlantic sector of the Southern Ocean. The sections and positions of stations at which DIC and TA samples were taken are indicated in Fig. 1, and were part of the following voyages (north to south): "Arctic Gateway" in 2008, 24.5° N in 2010, 24° S in 2009, and Drake Passages in 2009; Table 1 lists the nominal latitude/WOCE section, cruise name, year, ship, EXPO-code, and carbon PI. Hereafter, the cruises are referred to as DI332, DI346, JC032, and JC031.

All seawater samples for DIC and TA were drawn from the Niskin bottles of the Conductivity/Temperature/Depth (CTD) rosette into either 500 mL or 250 mL borosilicate ground glass bottles according to the Standard Operating Procedure (SOP) #1 (Dickson et al., 2007). In short, sample bottles are rinsed and then overfilled from the bottom. Subsequently during DI332, DI346, and JC032, samples were routinely poisoned by removing 1 % of sample volume and adding 0.02 % saturated mercuric chloride solution. Finally, the samples were stoppered with Apiezon-greased stoppers and stored in a cool, dark place, until analysed either onboard or back in the laboratory. During JC031, samples were only poisoned upon collection if the analysis did not take place

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within 8 h for surface samples and within 20 h for deep samples. Approximately 30 % of JC031 samples were poisoned.

DIC was analysed by coulometry (Johnson et al., 1985, 1987, 1993) following SOP #2 (Dickson et al., 2007) and TA by open cell potentiometric titration (Mintrop et al., 2000) following SOP #3b (Dickson et al., 2007). Two types of instruments were used: (i) for DIC only, a stand-along extraction unit (Robinson and Williams, 1991) connected to a coulometer (UIC, USA, model 5011), and (ii) for both DIC and TA, two Versatile INstrument for the Determination of Titration Alkalinity (VINDTA version 3C, Marianda, Germany, SN #004 and #007), connected to a coulometer (UIC, USA, model 5011) and a titrino (Metrohm UK Ltd.).

For the analysis, seawater is allowed into the stand-alone extractor by gravity, whilst it is drawn into the VINDTAs by slow peristaltic pump. Glass pipettes, approx. 20 mL for DIC and approx. 100 mL for TA, are first rinsed with new seawater sample before being filled to overflowing. The volumes of these pipettes were accurately calibrated before and after the cruises and the laboratory analysis.

The VINDTAs' sample pipettes and alkalinity titration cell are thermostated at 25 °C, and seawater samples were brought up to 25 °C prior to analysis. Conversely, the stand-alone extractor's DIC sample pipette is insulated but not water jacketed, and seawater samples were not warmed up prior to analyses.

During DIC analysis, all inorganic dissolved carbon is converted to CO_2 by addition of excess phosphoric acid (1M, 8.5 %) to a calibrated volume of seawater sample. Oxygen-free-nitrogen gas (OfN, BOC, UK), after passing through soda lime to remove any traces of CO_2, is used to carry the evolving CO_2 to the coulometer cell, where all CO_2 is quantitatively absorbed, forming an acid that is coulometrically titrated.

During TA analysis, aliquots of 0.1M hydrochloric acid is added to the seawater sample and the electromotive force is measured by a pH and reference electrode assembly. TA is calculated using a Gran plot and curve fit (Mintrop et al., 2000).

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A 500 mL bottle allows both DIC and TA to be analysed twice per sample (as a successive in-bottle duplicate), thereby providing information on the precision of measurements, whilst 250 mL bottles only allow DIC and TA to be analysed once.

The VINDTAs, and the stand-alone extractor during JC031 and JC032, were installed in a seagoing laboratory container of the Laboratory for Global Marine and Atmospheric Chemistry (LGMAC) of UEA, UK, on the ships' aft deck. During DI346, JC031, and JC032, DIC and TA analyses and 1st level quality control were performed at sea, and data were submitted, together with other cruise parameters, at end of cruise to the CLIVAR and Carbon Hydrographic Data Office (CCHDO). Samples were collected and stored during DI332 for VINDTA analysis back in the laboratory. Data submitted after 2nd level quality control are available at CCHDO, CDIAC, and are included in the GLODAP 2 effort.

3 Specific cruises' sampling and analyses

Table 2 lists the total number of stations sampled, depths sampled, and DIC and TA values reported for each cruise.

3.1 DI332, "Arctic gateway"

RRS Discovery cruise DI332, between 20 August and 25 September 2008 from Canada, via Greenland, to Scotland, was planned as one cruise in the vicinity of WOCE section AR7, as an occupation of both the western Labrador Sea part (AR7W) and the eastern part across the Irminger Basin, Iceland Basin, Rockall–Hatton Plateau and Rockall Trough (AR7E) (cruise track in Fig. 1a; Bacon, 2010). AR7W was completed, whilst AR7E could not be fully completed due to mechanical failures and foul weather.

A total of 74 CTD stations were occupied with a 24 × 20 L Niskin bottle rosette. West of Greenland (AR7W), Niskins of 28 stations were sampled into 250 mL bottles for DIC and TA analyses. Depths throughout the water column were sampled, with a minimum

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of three duplicate depths at each of the sampled stations (one set of duplicates at the deepest depth, one at the shallowest depth, and one set at an intermediate depth).

A total of 297 depths were sampled and stored for later analysis back in the laboratory, which was carried out between 30 July and 3 November 2009 using VINDTA SN #007. Instrument calibration was done throughout the analysis using CRMs (batch 90). Following 1st and 2nd level QC (see Sect. 4), 290 sample values of 21 stations were left, which contained 239 TA and 288 DIC values.

Second level quality controlled data have been submitted to BODC and to CCHDO, and are included in the GLODAP 2 effort, via CDIAC.

3.2 DI346, 24.5° N in 2010

RRS Discovery cruise DI346 was between 5 January and 19 February 2010 from Bahamas to Lisbon, Portugal, along nominal latitude 24.5° N in the Atlantic (cruise track in Fig. 1b; King and Hamersley, 2012). It was a repeat occupation of the section, previously occupied including carbon measurements in 2004 (e.g. Brown et al., 2010), 1998 (e.g. Macdonald et al., 2003), and 1992 (e.g. Rosón et al., 2003).

A total of 135 CTD stations were occupied with a 24 × 20 L Niskin bottle rosette. Samples for DIC and TA were not taken from all depths at each station. Generally, 16 depths were sampled from each station, including the shallowest and deepest ones with the other depths selected to allow for optimum interpolation across the whole section. Initially, all samples were taken in 500 mL bottles. From Station 34 until Station 129, every third station was fully sampled in 250 mL bottles and initially stored, with 4 depths of each of these stations sampled in duplicates bottles. All other stations, sampled in 500 mL bottles, were analysed as a priority and once profiles for these stations had been obtained, selected 250 mL bottles were analysed in order to strengthen areas of missing or suspect data.

Instrument calibration was done throughout the cruise using CRMs (batch 97), and 1st level quality controlled DIC and TA data were submitted, together with other cruise parameters, at end of cruise to the CLIVAR and Carbon Hydrographic Data Office

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(CCHDO), and 2nd level quality controlled data (1226 TA and 1322 DIC) have been included in the GLODAP 2 effort, via CDIAC.

3.3 JC032, 24° S in 2009

RRS James Cook cruise JC032 was between 7 March and 21 April 2009 from Montevideo, Uruguay, to Walvis Bay, Namibia, across the Brazil Current and along nominal latitude 24° S in the Atlantic Ocean (cruise track in Fig. 1c; King and Hamersley, 2010).

A total of 118 CTD stations were occupied with a 24 rosette of which 4 were 20 L Niskin bottles for near surface sampling and the remaining depths sampled using 10 L Niskin bottles. For DIC and TA, 116 stations were sampled. At each station sampled, the top two and bottom two were always sampled for DIC and TA, with in-between depths sampled alternatively for optimum interpolation across the whole section.

All samples were taken in 500 mL bottles with duplicate analyses DIC and TA done of each bottle, first DIC by the stand-alone extractor and VINDTA SN #007, followed by TA by the VINDTA SN #004 and #007 (the DIC on VINDTA SN #004 was inoperable during JC032).

Instrument calibration was done throughout the cruise using CRMs (bath 90), and 1st quality controlled DIC and TA data were submitted, together with other cruise parameters, at end of cruise to the CLIVAR and Carbon Hydrographic Data Office (CCHDO), and 2nd level quality controlled data (1504 TA and 1475 DIC) have been submitted to BODC and are included in the GLODAP 2 effort, via CDIAC.

3.4 JC031, Drake Passage in 2009

RRS James Cook cruise JC031, between 3 February and 3 March 2009 from Punta Arenas, Chile, to Montevideo, Uruguay, across Drake Passage (WOCE sections A21 and SR01b, cruise track in Fig. 1d; McDonagh and Hamersley, 2009). Section A21 (also known as SR01), had been occupied before in 1990 and 1999 with DIC measurements made in 1990 (Chipman et al., 1994), and section SR01b, located further to the east,

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has been occupied every year, except two, since 1993, with the 2009 occupation being the first time DIC and TA measurements were made here.

A total of 84 CTD stations were occupied with a 24-bottle Niskin bottle rosette with 10 L and 20 L bottles. For DIC and TA, a total of 63 stations were sampled. Samples for DIC and TA were taken from depths throughout the water column at each station, with a bias towards to top 1000 m.

All samples were taken in 500 mL bottles with duplicate analyses for DIC and TA done of each bottle, first DIC by the stand-alone extractor and VINDTA SN #007, followed by TA by the VINDTA SN #004 and #007 (the DIC part of VINDTA SN #004 was inoperable during JC031).

Instrument calibration was done throughout the cruise using CRMs (batch 90 and 92); data quality assurance and initial 1st level data quality control was done throughout the cruise, with full 1st level and 2nd level QC done post cruise. The data (1044 TA and 1060 DIC) have been included in the GLODAP 2 effort, via CDIAC.

4 Quality control

Quality control (QC) of DIC and TA measurements were done in 2 distinct steps: during 1st level QC, data are checked for obvious outliers, and technical or analytical problems during measurements; during 2nd level QC, cross-over analysis is performed with other sections and value corrections identified where necessary. Essentially, 1st level QC addresses precision whilst 2nd level QC addresses accuracy.

4.1 1st level QC

Throughout DIC and TA analyses, regular 500 mL Certified Reference Materials (CRMs) were analysed as in-bottle duplicates. Generally during one day's analyses, one CRM was run after the coulometer cell had stabilised, one mid-cell, and one at the end. This resulted in generally three, occasionally two CRMs being run per CTD cast.

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CRM batches used were #90 during DI332, #97 during DI346, #90 during JC032 and #90 and #92 during JC031.

Initial DIC and TA calibrations were done onboard (DI346, JC032, and JC031) or in the laboratory (DI332) by correcting all DIC and TA values by the difference between the mean of all CRM measurements and the certified CRM values of the respective batches used.

Quality flags of the World Ocean Circulation Experiment (WOCE, Joyce and Corry, 1994) were then assigned to each sample; when duplicates' flags were 2 or 3, the mean DIC or TA of the two was reported with the highest WOCE flag of the duplicates for DI346, JC032, and JC031.

4.2 2nd level QC

Second level QC was carried out using the Matlab crossover analysis toolbox (Tanhua, 2010). This method evaluates the consistency of deep-water measurements by comparing them with data from a reference dataset. The reference data used was the CARINA Atlantic dataset (updated August 2012) at http://cdiac.ornl.gov/ftp/oceans/2nd_QC_Tool/refdata/. Station profiles from the cruises in question were compared to profiles from cruises in the reference dataset which were within a certain horizontal distance (in this case 2° latitude). The result of each comparison between two cruises is an offset, which for TA and DIC is additive. For DI346 and JC032 data from below 1500 m depth were compared and the profiles were based on density whereas for DI332 the minimum depth was 1900 m and the profiles were based on depth (following Tanhua, 2010; Olsen et al., 2009). DI332 was compared with 14 cruises, DI346 with nine cruises, JC032 with two cruises and JC031 with two cruises. Corrective adjustments are only considered when offsets are greater than 4 $\mu mol\,kg^{-1}$ for DIC and 6 $\mu mol\,kg^{-1}$ for TA (Wanninkhof et al., 2003) and the mean offsets for the DIC and TA data were below these thresholds.

After QC, the accuracy (Table 3) of TA measurements was determined, defined as the standard deviation of CRMs per acid batch, and of DIC measurements as the

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standard deviation of all CRMs per cruise. The precision (Table 4) of measurements was determined, defined as the standard deviation of in-bottle duplicate measurements (Dickson et al., 2007, SOP 23).

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U. Schuster was PI of carbon sampling and analyses for DI332, JC032, and DI346. A. J. Watson was PI of the SOFI grant. D. C. E. Bakker was PI of carbon sampling and analyses for JC031. A. M. de Boer was watch keeper during JC032. E. M. Jones was watch keeper during JC031. G. A. Lee was watch keeper during DI346. O. Legge was watch keeper during DI346 and aided quality control of data. A. Louwerse was watch keeper during DI346. J. Riley was watch keeper during JC031. S. Scally was watch keeper during JC032.

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Table 1. Nominal latitude/WOCE section, cruise name, year, ship, EXPO-code, and carbon PI of DI332, DI346, JC032, and JC031.

Nominal latitude (WOCE section)	Cruise name	Year	Ship	EXPO code	Carbon PI
Arctic Gateway (AR7W and AR7E)	DI332	2008	*RRS Discovery*	74DI20080820	Ute Schuster
24.5° N (A05)	DI346	2010	*RRS Discovery*	74DI20100106	Ute Schuster
24° S (A09.5)	JC032	2009	*RRS James Cook*	74OH20090307	Ute Schuster
Drake Passage (A21 and SR01b)*	JC031	2009	*RRS James Cook*	74OH20090203	Dorothee Bakker

* The denotation A21/SR01 has been used both for cruises in the western Drake Passage (e.g. the western section on JC031 and 06MT11_5 (Chipman et al., 1994) or in the central Drake Passage e.g. RV *Laurence M. Gould cruise* LMG200603 (EXPO code 33LG20060321) and RV *Laurence M. Gould cruise* LMG200909 (EXPO code 33LG20090916)).

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Table 2. For DIC and TA, total number of (i) stations sampled, (ii) depths sampled, (iii) DIC and TA samples left after 1st and 2nd level quality control for each cruise.

Cruise name	Total number of			
	stations sampled	depths sampled	depths reported to data centres	
			of which TA values	of which DIC values
DI332	74	297	239	288
DI346	135	1427	1226	1322
JC032	116	1606	1504	1475
JC031	63	1380	1044	1060

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Table 3. Accuracy of DIC and TA measurements, defined as the standard deviation of CRM values around the mean, for TA as the mean of measurements for each acid batch.

	DIC accuracy [$\mu mol\,kg^{-1}$]			TA accuracy [$\mu mol\,kg^{-1}$]	
	VINDTA SN #004	VINDTA SN #007	Stand-alone extractor	VINDTA SN #004	VINDTA SN #007
DI332	NA	±2.1	NA	NA	±2.1
DI346	±3.0	±3.0	NA	±2.1	±3.0
JC032	NA	±3.1	±3.2	±3.2	±3.4
JC031	NA	±3.0	±3.0	±3.0	±3.0

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Table 4. Precision of DIC and TA measurements, defined as the standard deviation of measurements of in-bottle differences.

	DIC precision [$\mu mol\,kg^{-1}$]			TA precision [$\mu mol\,kg^{-1}$]	
	VINDTA SN #004	VINDTA SN #007	Stand-alone extractor	VINDTA SN #004	VINDTA SN #007
DI332	NA	±1.4	NA	NA	±0.9
DI346	±1.4	±1.8	NA	±1.2	±1.1
JC032	NA	±1.4	±1.7	±1.7	±1.8
JC031	NA	±2.7	±1.4	±2.3	±2.7

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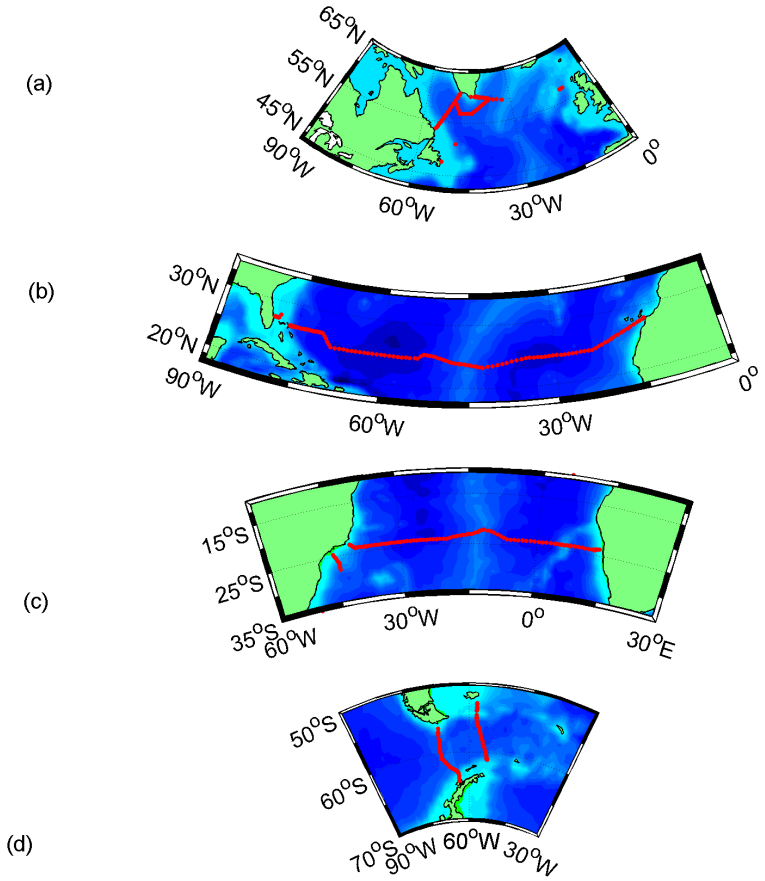

Fig. 1. Positions of stations at which DIC and TA samples were taken in the Atlantic and Drake Passage during **(a)** DI332 along "Arctic Gateway", **(b)** DI346 along 24.5° N, **(c)** JC032 along 24° S, and **(d)** JC031 across Drake Passage (A21 in the west and SR01b in the east).

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