Spatial variation of total CO₂ and total alkalinity in the northern Indian Ocean: A novel approach for the quantification of anthropogenic CO₂ in seawater

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ABSTRACT

As part of a cooperative effort of the Joint Global Ocean Flux Study (JGOFS) and of the World Ocean Circulation Experiment (WOCE) program, we have measured total CO₂ (TCO₂) and total alkalinity (TA) along three sections in the northern Indian Ocean. One section through the Gulf of Aden to the Arabian Sea is parallel to the coast of Yemen. One section is across the Arabian Sea along the nominal 9N latitude and the other section is across the Bay of Bengal along the nominal 10N latitude. The measurements were performed on board R/V *Knorr* in September-October 1995. The primary purpose of this work is to understand the penetration of anthropogenic CO₂ along these ocean sections. Here, we present a novel approach to the calculation of anthropogenic CO₂ in the ocean based upon the fundamentals of water-sources mixing. Consequently, we first describe the observations and mixing of water-sources before we describe the quantification of anthropogenic CO₂ concentrations in these waters. The data show large spatial variations in surface seawater of both total CO₂ (up to 50 μmol kg⁻¹) and total alkalinity (up to 40 μmol kg⁻¹). The variations are mainly associated with physical processes characterized by water masses of different temperature and salinity. For example, at depths we observed low TCO₂ concentration at longitude 54E ± 2E associated with the low-salinity water mass flowing northward. The contrasts between the sections across the Arabian Sea and the Bay of Bengal emphasize the large property differences between the two ocean basins. Multiparametric analyses on the data clearly show the relative contributions of different water-sources in each of the ocean sections. The mixing coefficients calculated from the multiparametric analyses are further used to quantify anthropogenic CO₂ concentrations in each water-source. The results indicate that the surface water-sources contain 47.8, 42.1 and 50.4 μmol kg⁻¹ in the Gulf of Aden, the Arabian Sea and the Bay of Bengal, respectively. In the surface waters there is slightly more anthropogenic CO₂ across the Bay of Bengal than across the Arabian Sea. In contrast, anthropogenic CO₂ has penetrated significantly deeper in the Gulf of Aden than in the Arabian Sea and in the Bay of Bengal.

1. Introduction

Since the GEOSECS program in the mid-70's, there have been relatively few measurements of the oceanic carbonate system in the northern Indian Ocean. This area is known to

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be a small CO_2 source for the atmosphere (Takahashi *et al.*, 1980; Batrakov *et al.*, 1981; Tans *et al.*, 1990). An early comparison between the data from the GEOSECS stations 417 ($12^\circ 59' \text{N}$, $64^\circ 27' \text{E}$) in the Arabian Sea and 446 ($12^\circ 30' \text{N}$, $84^\circ 29' \text{E}$) in the Bay of Bengal indicates that in January-March 1978, TCO_2 in surface seawater differed by $154 \mu\text{mol kg}^{-1}$ between these two ocean basins. The low TCO_2 concentrations and relatively cold temperatures observed at station 446 suggest that in contrast to the Arabian Sea, the Bay of Bengal is a sink for atmospheric CO_2 . However, very little is known about the temporal and spatial variations of carbon fluxes at the surface or at depths. Even less is known about the penetration of anthropogenic CO_2 in these ocean areas. Nevertheless, it is an intensely debated research subject. All current estimates of anthropogenic CO_2 concentrations in the ocean, including those based on the modification (Gruber *et al.*, 1996) of the original calculations (Brewer, 1978; Chen and Millero, 1979), are based upon several hypotheses such as the levels of pre-industrial pCO_2 in the atmosphere and in the surface ocean. Reasonable but non-negligible uncertainties are associated with these hypotheses. Here, we show that the good-quality data of the WOCE program can be used in a novel approach to reduce the uncertainties on the quantification of the anthropogenic CO_2 concentrations in the ocean.

The biogeochemical properties in surface waters of the North Indian Ocean are expected to vary seasonally since the strong seasonal variation of the surface ocean circulation are forced by the monsoons. Such seasonal variations of the carbon flux in the ocean and across the air-sea interface have recently been observed during the JGOFS program in the Arabian Sea (Millero *et al.*, 1998a; Goyet *et al.*, 1998c). However similar studies remain to be done in the Bay of Bengal.

Until recently there were very few TCO_2 and TA data available from these two basins. The GEOSECS data clearly indicated a significant difference between the properties of the Arabian Sea and the Bay of Bengal. Are the data of these few stations representative of typical water properties of the basins in which they were measured? One of the objectives of our work during the WOCEI1 cruise was to perform accurate measurements of TCO_2 and TA concentration to provide a detailed documentation of the water properties across these basins. The cruise occurred in September-October 1995, a different season from the observations during the GEOSECS program.

We present a detailed description of the differences in TCO_2 and TA measured during WOCE cruise I1 from the Gulf of Aden to the Arabian Sea and across the Bay of Bengal. The hydrologic, TA, and TCO_2 data are then analyzed to quantify the relative contributions of the water-sources to the observed concentrations. The results are used to quantify the concentrations of anthropogenic CO_2 in each of the water-sources and along the sections.

2. Sampling and analysis

The WOCE cruise I1 on R/V *Knorr* departed from Muscat (Oman) on August 29, 1995, and ended in Singapore on October 16, 1995. Three main sections were sampled; one in the Gulf of Aden, one from the coast of Somalia across the Arabian Sea to the shelf of India at a

nominal latitude of 9N, and a section from Sri Lanka across the Bay of Bengal to the Myanmar continental shelf at a nominal latitude of 10N (Fig. 1). There were a total of 158 hydrographic stations, three of which were stations reoccupied from either or both JGOFS and WOCE Cruise I7. Most of the stations were spaced 30' of longitude apart. A CTD/Rosette equipped with 36 conventional 10-1 Niskin bottles was used to collect samples at depth. We used 250-ml and 500-ml borosilicate glass bottles to collect subsamples for TCO₂ and TA measurements, respectively. The measurements were performed on board to eliminate contamination problems during storage and/or shipping.

All the equipment to measure total CO₂ and total alkalinity was shared by the CO₂ team of investigators (lead by Drs. A. Dickson, C. Goyet, R. Key, F. J. Millero, D. W. Wallace, and C. Winn) sponsored by the Department of Energy (DOE). The equipment remained on board R/V *Knorr* for the entire duration of the WOCE Indian Ocean Expedition (from January 1995 through January 1996). The sharing of equipment not only minimized shipping cost and set-up time, but further ensured that all the TCO₂ and TA data of the US WOCE Indian Ocean expedition were measured using the same analytical equipment with the same calibration procedures as described in the DOE handbook of methods for CO₂ analysis (DOE, 1994).

a. Total CO₂

TCO₂ was measured using two semi-automated multiparameter (SOMMA)/coulometric systems. The two systems were similar and used in parallel to process as many samples as possible. The principle of measurement as well as a complete description of the system used to perform accurate TCO₂ measurements can be found in the DOE handbook of methods for CO₂ analysis (DOE, 1994). The accuracy of the measurements, based on measurement of reference material provided by Dr. A. G. Dickson, was within a 2σ of ±4 μmol kg⁻¹ (Fig. 6 from Johnson *et al.*, 1998). A detailed assessment of the quality of the TCO₂ data from the Indian Ocean during the CO₂ survey cruises of the WOCE hydrographic program 1994–1996 is documented in Johnson *et al.* (1998).

b. Total alkalinity

Total alkalinity was measured by potentiometric acid titration (DOE, 1994). Here also, two identical systems were used in parallel to process as many samples as possible. The principle of measurement as well as a complete description of the system used to perform accurate TA measurements can be found in the DOE handbook of methods for CO₂ analysis (DOE, 1994). The accuracy of the measurements, based on measurement of reference material provided by Dr. A. G. Dickson, was generally within a 2σ of ±8.4 μmol kg⁻¹ (Fig. 4 from Millero *et al.*, 1998b). A detailed assessment of the quality of the TA data from the Indian Ocean during the CO₂ survey cruises of the WOCE hydrographic program 1994–1996 is documented in Millero *et al.* (1998b).

WOCE I1

Figure 1. Station locations of the cruise WOCE I1 (August 29–October 16, 1995) in the Northern Indian Ocean.

c. Partial pressure of CO_2

Partial pressure of CO_2 in surface seawater was continually measured by infrared analysis (Sabine and Key, 1996). Sabine and Key (1997) reported the details of these measurements and their associated accuracy during the CO_2 survey cruises of the WOCE hydrographic program 1994–1996.

3. Results

During the 49-day cruise, we collected and measured 2367 TCO_2 samples and 2367 TA samples. Examples of typical TCO_2 and TA-depth profiles are shown in Figure 2. This figure illustrates the typical low TCO_2 and TA concentrations in surface waters and their increase with depths. It also illustrates the large range of variations of TCO_2 concentrations compared with those of TA concentrations. In surface seawater, TCO_2 and TA concentrations in the Bay of Bengal (station 1004) are significantly lower than those in the Arabian Sea (station 908).

Figure 3, 4, and 5 illustrate the concentration of TCO_2 and TA, along each of the three sections of the cruise. Along the nominal latitude of 9N, TCO_2 in surface seawater is highest ($2020 \mu\text{mol kg}^{-1}$) on the eastern side of the Arabian Sea (Fig. 3) and is lowest (below $1850 \mu\text{mol kg}^{-1}$) on the eastern side of the Bay of Bengal (Fig. 4). The difference ($>170 \mu\text{mol kg}^{-1}$) between these observations is larger than typical TCO_2 seasonal variations in other ocean areas (less than $100 \mu\text{mol kg}^{-1}$), larger than the anthropogenic signal (in the order of $50 \mu\text{mol kg}^{-1}$), and larger than most spatial variations over similar distances. At depth, the cross-section of TCO_2 in the Arabian Sea clearly shows the low TCO_2 water coming from the south along longitude $54\text{E} \pm 2^\circ$. This deep boundary current between the Arabian Basin and the northern Somali Basin flows through the Owen Fracture Zone at depth 3500–4000 m (Johnson *et al.*, 1991a,b).

In contrast, the highest TA concentrations in surface seawater ($>2400 \mu\text{mol kg}^{-1}$) were observed around 66E longitude in the Arabian Sea, and the lowest TA concentrations ($<2140 \mu\text{mol kg}^{-1}$) were observed on the eastern side of the Bay of Bengal. The maxima and minima were associated with the maxima and minima in salinity (Fig. 6). These observations confirm that TA in surface seawater is linearly correlated with salinity (Brewer *et al.*, 1986; Takahashi *et al.*, 1980; Millero *et al.*, 1998c). The geometric mean correlation (Ricker, 1973; Laws and Archie, 1981), between salinity (S) and total alkalinity (TA) in surface waters including all surface data from the WOCE II cruise follows the relationship:

$$\text{TA} = 350.8 + 55.6 \text{ S } (\pm 5.7) \quad (31 < \text{S} < 36.8) \quad (R^2 = 0.993) \quad (1)$$

At the time of the measurement, TCO_2 in surface water along the section from the Gulf of Aden displayed significant spatial variations reflecting the various water masses (Fig. 5). For instance, the data indicate a maximum of $2175 \mu\text{mol kg}^{-1}$ at station 889 ($14^\circ 30'\text{N}$, $50^\circ 50'\text{E}$) and a minimum of $1999 \mu\text{mol kg}^{-1}$ at station 881 (13°N , $48^\circ 20'\text{E}$). Over the section, TCO_2 in surface water increased eastward. In contrast, total alkalinity in surface

Figure 2. Observations of TCO₂ and TA concentrations(μmol kg⁻¹) in the Arabian Sea at station 908 (8.50N, 54.22E) and in the Bay of Bengal at station 1004 (9.97N, 93.22E).

water increased westward. Below 300 m, the high total alkalinity concentration in the Gulf of Aden is associated with high salinity (Fig. 6a). This is a characteristic of Red Sea water entering into the Arabian Sea.

Figure 7 illustrates the variations of pCO₂ in surface seawater (pCO₂^{sea}) and in the atmosphere (pCO₂^{atm}) along the three sections. At the time of the measurement (late August-early September), the Gulf of Aden was a source of CO₂ for the atmosphere with a ΔpCO₂ (pCO₂^{atm} – pCO₂^{sea}) in the order of +50 μatm. Across the Arabian Sea in September at nominal latitude of 9N, ΔpCO₂ decreased approximately from +50 to

Figure 3. Total CO₂ ($\mu\text{mol kg}^{-1}$) (a), and total alkalinity ($\mu\text{mol kg}^{-1}$) (b), across the Arabian Sea along the nominal 9N latitude. Crosses on the TCO₂ and TA sections show the location of the measured samples. Note the changes of coordinates and isoline spacing in the paired panels.

Figure 4. Total CO₂ ($\mu\text{mol kg}^{-1}$) (a), and total alkalinity ($\mu\text{mol kg}^{-1}$) (b), across the Bay of Bengal along the nominal 10N latitude. Crosses on the TCO₂ and TA sections show the location of the measured samples. Note the changes of coordinates and isoline spacing in the paired panels.

Figure 5. Total CO₂ ($\mu\text{mol kg}^{-1}$) (a), and total alkalinity ($\mu\text{mol kg}^{-1}$) (b), from the Gulf of Aden to the Arabian Sea. Crosses on the TCO₂ and TA sections show the location of the measured samples. Note the changes of coordinates and isoline spacing in the paired panels.

Figure 6. Salinity along sections (a) from the Gulf of Aden to the Arabian Sea, (b) in the Arabian Sea, and (c) across the Bay of Bengal.

Figure 7. Partial pressure of CO_2 in surface seawater (circles) and in the atmosphere (triangles) (a) from the Gulf of Aden to the Arabian Sea, (b) in the Arabian Sea, and (c) across the Bay of Bengal.

zero μatm from the Somali coast to the Indian coast, respectively. This is in good agreement with the estimated seasonal variations of $\text{pCO}_2^{\text{sea}}$ in the Arabian Sea based on the observations from the JGOFS cruises in 1995 (Goyet *et al.*, 1998c). In contrast to the Arabian Sea and the Gulf of Aden, ΔpCO_2 (in late September-early October) across the Bay of Bengal remained close to zero ($-10 \mu\text{atm} < \Delta\text{pCO}_2 < +10 \mu\text{atm}$). The observations of $\text{pCO}_2^{\text{sea}}$ are in good agreement with the observations of TCO_2 and TA in surface

Figure 8. Ratio of TCO₂ and TA along sections (a) in the Arabian Sea, and (b) across the Bay of Bengal.

seawater. Overall the data suggest that in August/September the North Indian Ocean was a small source of CO₂ for the atmosphere.

The pCO₂ in seawater is dependant upon seawater temperature and the ratio TCO₂/TA (Copin-Montegut, 1988; Goyet *et al.*, 1993). Seawater pCO₂ is more sensitive with temperature when the ratio (TCO₂/TA) is closer to one than zero. Figure 8 shows a

comparison of the TCO_2/TA ratio along the Arabian Sea section with that along the Bay of Bengal section. In surface water, this ratio is highest (~ 0.87) close to the Somali coast, and is lowest (< 0.84) between longitudes 68E and 72E. This small difference (0.03) in the TCO_2/TA ratio corresponds to a pCO_2 difference of approximately 18 μatm for a five-degree change in water temperature (Goyet *et al.*, 1993). On the other hand, the ratio remains close to 0.85 across the Bay of Bengal. As a result, a small change in surface temperature close to the Somali coast up to 52E will have a larger effect on pCO_2 than a similar temperature change at longitude 69E $\pm 3^\circ$ or across the Bay of Bengal. This TCO_2/TA ratio further illustrates the differences in the water properties in these two ocean basins. The 0.96 isoline of this ratio is located between 150 m and 200 m across the Bay of Bengal while it increases eastward across the Arabian Sea from 400 m to 250–200 m.

4. Relative contribution of different water-sources to the observed temperature, salinity, oxygen, and total alkalinity concentrations

Based upon the original work of two-end member water mixing by Broecker (1974) and Broecker and Peng (1982), we used a multi-parameter analysis (Mackas *et al.*, 1987; Tomczak, 1981) to quantify the contributions of the different water sources to the observed data. The analysis is based on an inverse method that uses data from tracers to estimate the relative importance of each water-source to the composition of the observed water. Many applications, at regional or basin scales, have been undertaken in several oceanic regions (Mackas *et al.*, 1987; Tomczak and Large, 1989; Maamaatuaiahutapu *et al.*, 1992; Hinrichsen and Tomczak, 1993; You and Tomczak, 1993; You, 1997). The approach was also recently used to quantify the water mass distribution in the Indonesian throughflow entering the Indian Ocean (Coatanoan *et al.*, 1999). In this latter study, nutrients were used as nonconservative tracers and additional terms were added to the equations to take into account the variation of nutrient concentrations due to biological activity. Here, we use temperature and salinity as conservative tracers, and oxygen and total alkalinity as nonconservative tracers. Since a detailed description of the multiparametric analysis is provided by many of the early work (Mackas *et al.*, 1987; Tomczak and Large, 1989; You and Tomczak, 1993), only a brief description is given below.

The conservation equation for the conservative tracers (temperature and salinity) can be expressed as follows:

$$C_m = \sum_{j=1}^n k_j C_j \quad (2)$$

where k_j represents the mixing coefficient of the water-source j , n is the number of water-sources, C_j is the conservative tracer concentration of the water-source j , and C_m is the measured tracer concentration of the seawater sample.

The conservation equation for the nonconservative tracers (oxygen and total alkalinity)

can be expressed as follows:

$$NC_m = \sum_{j=1}^n k_j NC_j + \Delta NC \quad (3)$$

where k_j represents the mixing coefficient of the water-source j , n is the number of water-sources, NC_j is the nonconservative tracer concentration of the water-source j , ΔNC represents an unknown biogeochemical contribution to the tracer NC , and NC_m is the measured concentration of the nonconservative tracer in seawater.

Adding to the previous equations, we use a strict equation to provide mass conservativity: $\sum k_j = 1$. Convex constraints that express the non-negativity of the coefficient, $k_j \geq 0$ for all j , are also added to the matricial system resolved by the inverse method. The stability of the results is tested by numerical perturbation experiments. These experiments consist of adding random deviations to the measured data and to the water-source tracers. The random deviations are calculated for each tracer and for each perturbation from a relative error (Metzl *et al.*, 1990). These relative errors provide larger deviation on the surface water-source than deep water-sources. Thirty perturbations at each station give a mean solution for each mixing coefficient and the study of standard deviations leads to evaluate the coefficient validity. To get information about nonconservative behavior of tracers and possible errors due to the water-source description and to uncertainties associated with the measurements, a vector of error on the tracer is calculated. This vector corresponds to the difference between the predicted and the measured data.

The different sources of water masses are typically identified using the two conservative tracers temperature and salinity by analyzing the θ - S diagrams. Here we have considered the diagrams of the oxygen and total alkalinity tracers versus the hydrological tracers (temperature and salinity) as well as details on Indian water masses given by previous studies (Emery and Meincke, 1986; Wyrki, 1971; You, 1997; You and Tomczak, 1993). Along the cruise track, the θ - S diagram (Fig. 9) indicates that there are eight different water-sources; the Red Sea surface water (RSSW), the Arabian Sea surface water (ASW), the Bay of Bengal surface water (BBSW), the Red Sea intermediate water (RSW), the Indian intermediate water (IIW), the Bay of Bengal intermediate water (BBIW), the deep Indian water (DIW), and the bottom water (BW). Using more than the two conservative tracers (temperature and salinity) and taking into account some constraints in the multiparametric analysis, all these eight water-sources are preserved for the analysis.

The Red Sea intermediate water (RSW) is characterized by a salinity maximum. It spreads into the Arabian Sea between 500 and 800 m (Wyrki, 1971). In surface, high salinity of the Red Sea surface water (RSSW) is due to the high evaporation developed in this region (Premchand *et al.*, 1986). The intermediate layer in the Arabian Sea is a mixing between the highly-saline water of the Red Sea and the Persian Gulf. A typical feature of the Arabian Sea intermediate waters is the scarcity of oxygen (Eremeev *et al.*, 1994; Wyrki, 1971). However, a weak oxygen maximum is observed in the southwestern

Figure 9. θ -S diagram of the WOCE 11 cruise. The black lines, dashed lines and grey lines respectively correspond to the Gulf of Aden, Arabian Sea, and Bay of Bengal sections. The water-sources are indicated by white circles (BBSW: Bengal Bay Surface Water, ASW: Arabian Sea surface Water, RSSW: Red Sea Surface Water, IIW: Indian Intermediate Water, BBIW: Bengal Bay Intermediate Water, RSW: Red Sea Water, DIW: Deep Indian Water, BW: Bottom Water).

Arabian Sea. We will call this water the Indian intermediate water (IIW). The surface water in the Bay of Bengal is subject to high precipitation and output of rivers. Consequently, low salinity characterizes the Bay of Bengal surface water (BBSW). Deeper, the Bay of Bengal intermediate water (BBIW) is represented by higher salinity and low oxygen contents. Below the intermediate waters, the deep Indian water (DIW) and the bottom water (BW) are characterized by high-oxygen.

The tracer water-source characteristics are summarized in Table 1. The characterization of these water sources is typically led by a cluster analysis (You and Tomczak, 1993). As a result, isolated extreme points due to local processes (river output, . . .) cannot be taken into account to characterize water sources which in fact are not "water sources." The seasonal study of the water properties (Wyrтки, 1971; Goyet *et al.*, 1998b) indicates that the seasonal

Table 1. Tracer characteristics in each water-source determined from the WOCE I1 data. The sources are: Bengal Bay Surface Water (BBSW), Bengal Bay Intermediate Water (BBIW), Red Sea Surface Water (RSSW), RSW (Red Sea Water), ASW (Arabian Sea Water), Indian Intermediate Water (IIW), Deep Indian Water (DIW), and Bottom Water (BW).

	θ (°C)	Salinity	Oxygen ($\mu\text{mol kg}^{-1}$)	TA ($\mu\text{mol kg}^{-1}$)	TCO_2^{ws} ($\mu\text{mol kg}^{-1}$)
BBSW	29.0	32.80	220	2180	1853
BBIW	12.5	35.02	18	2310	2219
RSSW	29.0	36.30	220	2390	2052
RSW	13.5	36.70	20	2360	2241
ASW	29.0	35.30	220	2310	1989
IIW	13.0	35.20	90	2320	2193
DIW	1.8	34.75	156	2445	2331
BW	0.5	34.72	200	2410	2297

variability of the sea surface properties in the studied area is not large enough to meaningfully change the results of the mixing coefficients.

The tracer residuals (Fig. 10) are characterized by typically low values throughout the water column compared to the large range of the water-source properties in the northern Indian Ocean. The largest (negative) residuals are observed in the upper layer for oxygen. These residuals are due to the air-sea interactions that are not considered in the multiparametric analysis.

The results of these analyses for the intermediate waters (RSW, IIW and BBIW) and the deep and bottom water (DIW and BW) are shown in Figures 11 and 12. Only the most representative distributions are presented. The results indicate high mixing proportions (k_j) of the Red Sea water in the Gulf of Aden (Fig. 11a) which decrease rapidly during its spreading to the Arabian Sea (Fig. 11c). The core of that water (RSW) is centered between 500 m and 1200 m (upper and lower boundaries, respectively). The RSW properties are transformed by strong vertical mixing in the Arabian Sea. The intermediate water in the Arabian Sea spreads over a large fraction of the water column (250–1500 m). A mixing of five water-sources, namely RSW, IIW, BBIW, DIW, and BW (Fig. 11b–f) constitute it. The contribution of each of these water masses is similar (approximately 20%), due to the strong mixing. The deep and bottom water below 1500 m (Fig. 11e,f), incorporate the highly-saline Arabian Sea waters and transformed Antarctic bottom waters. They largely participate in the mixing of the upper-layer waters.

In the Bay of Bengal (Fig. 12c–f), the mixing of the BBIW, DIW, and BW constitutes the intermediate layer. The mixing proportions of RSW and IIW (Fig. 12b,c) are low. Their participation in the mixing water of the Bay of Bengal is not significant. In comparison to the Arabian Sea, the deep and bottom waters (DIW and BW) also participate in the mixing of the upper-layer waters (Fig. 12e,f).

The distributions of biogeochemical unknowns (ΔO_2 and ΔTA) are presented in

Figure 10. Residuals of (a) potential temperature ($^{\circ}C$), (b) salinity, (c) oxygen ($\mu mol\ kg^{-1}$), and (d) total alkalinity ($\mu mol\ kg^{-1}$) for all sections of the WOCE II cruise.

Figure 13. They indicate high values of oxygen consumption in the upper layers. The consumption is more important in the Bay of Bengal (Fig. 13c) than in the Arabian Sea (Fig. 13b). There is also a significant difference between the eastern and western side of the Arabian Sea section. Higher consumption in the eastern side is the consequence of the productive area near the coast of India during this season (Kuzmenko, 1994). Low values at the depth of the Red Sea Water along the Gulf of Aden section (Fig. 13a), indicate that

Figure 11. Mixing proportions of (a) RSW (Red Sea Water) in the Gulf of Aden, (b) IIW (Indian Intermediate Water), (c) RSW (Red Sea Water), (d) BBIW (Bengal Bay Intermediate Water), (e) DIW (Deep Indian Water), and (f) BW (Bottom Water) in the Arabian Sea.

this water has relatively high oxygen content prior to mixing with the Arabian Sea water. The decrease of ΔO_2 values at depths is due to the advection of oxygenated Antarctic waters. The distribution of ΔTA (Fig. 13d-f) indicates negative values in the upper layer and positive values in the lower layer. These variations result from formation/dissolution of calcium carbonate and respiration (Goyet and Brewer, 1993). Note that in practice both ΔO_2 and ΔTA signals also include measurement uncertainties.

Figure 12. Mixing proportions of (a) BW (Bottom Water) in the Gulf of Aden, (b) IIW (Indian Intermediate Water), (c) RSW (Red Sea Water), (d) BBIW (Bengal Bay Intermediate Water), (e) DIW (Deep Indian Water) and (f) BW (Bottom Water) in the Bay of Bengal.

5. Estimation of anthropogenic CO₂ in seawater from the multiparametric analysis

The variations of TCO₂ in seawater can be represented by the equation:

$$\text{TCO}_2 = \text{TCO}_2^{\text{cir}} + \text{TCO}_2^{\text{bio}} + \text{TCO}_2^{\text{ant}} \quad (4)$$

where TCO₂ represents TCO₂ concentration at the considered location,

Figure 13. Distribution of ΔO_2 ($\mu\text{mol kg}^{-1}$) and ΔTA ($\mu\text{mol kg}^{-1}$) along the ocean sections (a) and (d) in the Gulf of Aden, (b) and (e) across the Arabian Sea and, (c) and (f) across the Bay of Bengal.

$\text{TCO}_2^{\text{cir}}$ represents the TCO_2 concentration resulting from mixing the different water masses by ocean circulation,

$\text{TCO}_2^{\text{bio}}$ represents the contribution of TCO_2 concentration due to biological processes,

$\text{TCO}_2^{\text{ant}}$ represents the contribution of TCO_2 concentration due to the penetration of anthropogenic CO_2 .

Below the mixed layer depth $\text{TCO}_2^{\text{bio}}$ can be parameterized as a function of oxygen and total

alkalinity according to the equation (Brewer, 1978):

$$\text{TCO}_2^{\text{bio}} = a\Delta\text{O}_2 + 0.5 \Delta\text{TA} \quad (5)$$

where the coefficient “ a ” depends upon the Redfield ratio (Redfield, 1934). This coefficient is estimated to be in the order of 0.78 ± 0.04 (Brewer, 1978; Goyet and Brewer, 1993; Brewer *et al.*, 1997; Goyet *et al.*, 1998a).

ΔO_2 represents the variation in oxygen due to biological activity. In practice, ΔO_2 is often assimilated with the Apparent Oxygen Utilization (AOU; difference between the O_2 concentration of the atmospherically equilibrated seawater (T, S) and the observed O_2 concentration). The comparison between ΔO_2 calculated from the multiparametric analysis and AOU indicates a significant correlation:

$$\text{AOU} = 148.5 - 1.37 \Delta\text{O}_2 \quad (R^2 = 0.75) \quad (z \geq 200 \text{ m}) \quad (6)$$

Since ΔO_2 varies between -20 and -80 , AOU is approximately a factor 3.5 larger than ΔO_2 .

ΔTA represents the variation in total alkalinity. In practice ΔTA is often calculated as the difference between the observed TA concentration and the TA concentration this water had (TA_0) when it was at the surface and in contact with the atmosphere. TA_0 is often estimated as a linear function of salinity (Brewer *et al.*, 1986; 1997; Millero *et al.*, 1998c). Since TA_0 is not affected by the penetration of anthropogenic CO₂, ΔTA is generally close to the observed difference between TA measured at depth and TA in surface seawater. This difference below 2000 m is in the order of 100 to 200 $\mu\text{mol kg}^{-1}$ depending upon the location (Fig. 2). The comparison between ΔTA calculated using an estimated TA_0 and ΔTA calculated from the multiparametric analysis also indicates that the former ΔTA is higher by approximately a factor of 4 than the latter.

We calculated $\text{TCO}_2^{\text{bio}}$ in this work using $a = 0.78$ and the results of the multiparametric analysis for the values of ΔO_2 and ΔTA . There is no assumption about the apparent oxygen utilization or the initial concentration of TA. The calculated ΔO_2 and ΔTA (Fig. 13) are the direct result of the difference between the observed concentrations and the concentrations calculated from the mixing of the different water-sources. Note that because ΔO_2 and ΔTA values calculated using the multiparametric analyses are approximately 3.5 and 4 times respectively lower than AOU and the previous determination of ΔTA , $\text{TCO}_2^{\text{bio}}$ calculated here is close to that calculated using the method first described by Brewer (1978).

Now, imagine for a moment that we had also observed the ocean at a pre-industrial time. The TCO_2 concentrations in the water sources $[\text{TCO}_2^0]_j$ at that time would not contain any anthropogenic CO₂. Since the mixing coefficients k_j are known, the contemporary $\text{TCO}_2^{\text{ant}}$ could be estimated below the mixed layer depth from the present data and the pre-industrial

Figure 14. Residuals between the observed and calculated TCO_2 data calculated from Eq. 8 using the WOCE II data throughout the water column below 200 m: (a) with $F_j(z) = 1$. The mean and standard deviation are -2.3 and $7.9 \mu\text{mol kg}^{-1}$ respectively (b) with $F_j(z) = e^{-((z-z_0)/H)}$. The mean and standard deviation are 0.4 and $5.2 \mu\text{mol kg}^{-1}$ respectively (c) with $F_j(z) = a - b/[1 + e^{-(c(z-d))}] + b$. The mean and standard deviation are 0.02 and $5.7 \mu\text{mol kg}^{-1}$ respectively. For comparison the standard deviation of the TA and TCO_2 measurements are within 8.4 and $4 \mu\text{mol kg}^{-1}$, respectively.

$[\text{TCO}_2^0]_j$ using the equation:

$$\text{TCO}_2 - (0.78 \Delta\text{O}_2 + 0.5 \Delta\text{TA}) = \sum_{j=1}^n k_j [\text{TCO}_2^0]_j + \text{TCO}_2^{\text{ant}}. \quad (7)$$

In reality, anthropogenic CO_2 has penetrated into the ocean since pre-industrial time, hence, the concentrations $[\text{TCO}_2^0]_j$ are not known (except in deep waters). Since the properties of the water-sources (Table 1) were defined from our 1995 data, the TCO_2 concentrations in surface and intermediate water sources $[\text{TCO}_2^{\text{ws}}]_j$ include anthropogenic CO_2 concentrations:

$$[\text{TCO}_2^{\text{ws}}]_j = [\text{TCO}_2^0]_j + [\text{TCO}_2^{\text{ant}}]_j. \quad (8)$$

Thus, Eq. 7 can be written:

$$\text{TCO}_2 - (0.78 \Delta\text{O}_2 + 0.5 \Delta\text{TA}) = \sum_{j=1}^n k_j [[\text{TCO}_2^0]_j + [\text{TCO}_2^{\text{ant}}]_j F_j(z)] \quad (9)$$

where $F_j(z)$ are decreasing functions (from one to zero) with depth z . A function $F(z)$ is introduced here to reflect the decrease of anthropogenic concentration with the increasing age (and consequently depth) of the waters. Note that if $F_j(z) = 1$, the residuals (Fig. 14a) of Eq. 9 have a marked minimum at approximately 1000 m. This minimum is associated with the erroneous overestimated mixing of anthropogenic CO_2 from today's surface

waters that in reality have not yet reached this depth. The functions $F_j(z)$ are necessary to represent the time dependence of the penetration of anthropogenic CO₂ in seawater. Since these functions should reflect the slow penetration of anthropogenic CO₂ at depths in each water-source, we investigated two functions; an exponential function (Papaud and Poisson, 1986) of the form:

$$F_j(z) = e^{-((z-z_0)/H)} \quad (10)$$

and a sigmoid function (Goyet and Davis, 1997) of the form:

$$F_j(z) = a - b/[1 + e^{-(c(z-d))}]^e + b. \quad (11)$$

The exponential functions (Eq. 10) have the property of decreasing sharply from their maximum at depths just below their maximum. In contrast, the sigmoid functions (Eq. 11) can decrease at depths significantly deeper than the depth of their maximum. We show in our earlier work (Goyet and Davis, 1997), that sigmoid functions are indeed best suited to represent TCO₂ data in the upper ocean.

Note that these functions $F_j(z)$ do not introduce the assumption that anthropogenic CO₂ is mixing vertically. If it were so, only one function would be used. The four different sigmoid functions used here take into account the time and thus the penetration of anthropogenic carbon along density surfaces. The “step” shape of the function takes into account the change from a density surface to another. Only these sigmoid functions (in contrast to other constant, linear, or exponential functions) minimize the residuals of the equation randomly around zero. Thus indicating that these sigmoid functions can be used as a proxy to correctly represent the process (whatever it might be) of penetration of anthropogenic CO₂ in the ocean.

Below 200 m, Eq. 9 is solved, for the two sets of unknowns $[\text{TCO}_2^s]_j$ and $[\text{TCO}_2^{\text{ant}}]_j$, with Eq. 8 as constraint and an initial guess of 45 $\mu\text{mol kg}^{-1}$ for $\text{TCO}_2^{\text{ant}}$ in surface seawaters and 20 $\mu\text{mol kg}^{-1}$ for $\text{TCO}_2^{\text{ant}}$ in intermediate waters. These values correspond to the values that minimize the residuals (Fig. 14). The 0–200 m layer is not used for this analysis since Eq. 5 and consequently Eq. 9 is not valid in the upper layer due to biological activity and air-sea exchange (as seen in Fig. 10c) that are not considered in the equation.

Four functions $F_j(z)$ (Table 2) were introduced according to the region (Bay of Bengal and Arabian Sea-Red Sea) and to the water layer (surface and intermediate waters). The variables of these functions are calculated iteratively to minimize the residuals of Eq. 9. The residuals of Eq. 9 are significantly smaller and more randomly distributed around zero when the functions $F_j(z)$ are sigmoid functions (Fig. 14c) than when the $F_j(z)$ are exponential functions (Fig. 14b). The mean and standard deviation (2σ) of all the residuals are 0.02 and 5.7 $\mu\text{mol kg}^{-1}$ respectively for the application with the sigmoid functions.

The results (Table 3) indicate that the concentration of anthropogenic CO₂ in the surface water-source of the Red Sea, Arabian Sea, and Bay of Bengal are 48 $\mu\text{mol kg}^{-1}$, 42 $\mu\text{mol kg}^{-1}$, and 50 $\mu\text{mol kg}^{-1}$, respectively. These results further show that the constraints (Eq. 8) are not exactly respected. This reflects the fact that the observed

Table 2. Estimated constants for the sigmoid and exponential function according to the region and the water layer.

	Sigmoid function $-F(z)$					Exponential function $F(z)$			
						$Z < 600\text{ m}$		$z > 600\text{ m}$	
	a	b	c	D	e	z_0	H	z_0	H
Bay of Bengal Surface layer	0	-1	0.006	150	9	40	200	40	200
Bay of Bengal Intermediate layer	0	-1	0.012	700	9	$F(z) = 1$		600	340
Arabian Sea-Red Sea Surface layer	0	-1	0.0065	70	9	30	100	30	100
Arabian Sea-Red Sea Intermediate layer	0	-1	0.018	800	9	$F(z) = 1$		700	320

concentrations at the time of the measurement may not be exactly the annual mean concentrations of the water-sources. As expected from Eq. 9, the results for $\text{TCO}_2^{\text{ant}}$ in the surface water-sources are more sensitive than for $\text{TCO}_2^{\text{ant}}$ in the intermediate water-sources to the values ($45\text{ }\mu\text{mol kg}^{-1}$ and $20\text{ }\mu\text{mol kg}^{-1}$) of the initial guess. However, since as mentioned above, these initial values for $\text{TCO}_2^{\text{ant}}$ are chosen iteratively to minimize the residuals of Eq. 9, the uncertainties introduced by the choice of these initial values are minimal.

The concentration of anthropogenic CO_2 in seawater along these sections can then be computed throughout the water column from the equation:

$$\text{TCO}_2^{\text{ant}} = \sum_{j=1}^n k_j [\text{TCO}_2^{\text{ant}}]_j F_j(z).$$

(12)

Figure 15 illustrates the results of Eq. (12) applied (using the sigmoid functions) along the three sections. The $5\text{ }\mu\text{mol kg}^{-1}$ isoline is around 1500 m in the section of the Gulf of Aden. It is slightly below 1000 m across the Arabian Sea and it is slightly above 1000 m across the Bay of Bengal. These results indicate that anthropogenic CO_2 has penetrated significantly deeper in the Gulf of Aden than in the Arabian Sea and in the Bay of Bengal.

Table 3. Initial TCO_2° and anthropogenic $\text{TCO}_2^{\text{ant}}$ concentrations for each water-source.

	TCO_2° ($\mu\text{mol kg}^{-1}$)	$\text{TCO}_2^{\text{ant}}$ ($\mu\text{mol kg}^{-1}$)
BBSW	1786	50.4
BBIW	2198	4.2
RSSW	2012	47.8
RSW	2243	9.6
ASW	1960	42.1
IIW	2170	13.9
DIW	2322	—
BW	2297	—

Figure 15. Anthropogenic CO₂ concentration along ocean sections (a) in the Gulf of Aden, (b) across the Arabian Sea, (c) across the Bay of Bengal.

In contrast, in the surface waters there is slightly more anthropogenic CO₂ across the Bay of Bengal than across the Arabian Sea.

6. Estimation of the accuracy of the quantification of anthropogenic CO₂ concentrations

The accuracy of anthropogenic CO₂ concentration can be determined from the accuracy associated with the TCO₂ measurement (σ_{TCO_2}), the coefficient “ a ” from Eq. 5 (σ_a), the variations in oxygen and total alkalinity ($\sigma_{\Delta\text{O}_2}$ and $\sigma_{\Delta\text{TA}}$), the TCO₂⁰ derived from the water-source characterization ($\sigma_{\text{TCO}_2^0}$), and the accuracy associated with the mixing coefficients (σ_k). We assume that the uncertainties associated with the sigmoid functions are negligible. This assumption is reasonable since these functions vary between zero and one and have less influence on the results as they approach zero. Finally, the error of TCO₂^{ant}, ($\sigma_{\text{TCO}_2^{\text{ant}}}$), is given by:

$$\begin{aligned}\sigma_{\Delta\text{TCO}_2^{\text{ant}}}^2 = & \{(\delta\text{TCO}_2^{\text{ant}}/\delta\text{C}) \sigma_{\text{TCO}_2}\}^2 + \{(\delta\text{TCO}_2^{\text{ant}}/\delta\Delta\text{O}_2) \sigma_{\Delta\text{O}_2}\}^2 \\ & + \{(\delta\text{TCO}_2^{\text{ant}}/\delta a) \sigma_a\}^2 + \{(\delta\text{TCO}_2^{\text{ant}}/\delta\Delta\text{TA}) \sigma_{\Delta\text{TA}}\}^2 \\ & + \{(\delta\text{TCO}_2^{\text{ant}}/\delta k) \sigma_k\}^2 + \{(\delta\text{TCO}_2^{\text{ant}}/\delta\text{TCO}_2^0) \sigma_{\text{TCO}_2^0}\}^2\end{aligned}\quad (13)$$

where $\sigma_{\text{TCO}_2} = 2 \mu\text{mol kg}^{-1}$, $\sigma_{\Delta\text{O}_2} = 2 \mu\text{mol kg}^{-1}$, $\sigma_a = 0.04$, $\sigma_{\Delta\text{TA}} = 4 \mu\text{mol kg}^{-1}$, $\sigma_k = 0.05$ and $\sigma_{\text{TCO}_2^0} = 2 \mu\text{mol kg}^{-1}$. The values for these uncertainties were derived according to the uncertainties of the measurement, sensitivity study of the mixing coefficient, and standard deviations of biological parameters (ΔO_2 and ΔTA). The uncertainty σ_a has been determined by several authors (Brewer, 1978; Goyet and Brewer, 1993; Brewer *et al.*, 1997; Goyet *et al.*, 1998a). The uncertainty of the coefficient 0.5 of the Eq. (5) is too low to significantly introduce errors on the anthropogenic CO₂ quantification. The standard deviations on the mixing coefficients are the highest where these coefficients are the lowest and consequently do not participate in the determination of the anthropogenic CO₂. The uncertainty σ_k is taken from the average of the standard deviation values observed where the water sources are significantly represented with their highest associated coefficients. The uncertainties on TCO₂^{ant} due to the uncertainties on the mixing coefficient are estimated $< 2.0 \mu\text{mol kg}^{-1}$. In addition, since most of the anthropogenic CO₂ will be found in waters with relatively low ΔO_2 ($< 50 \mu\text{mol kg}^{-1}$), the sum of the uncertainties for TCO₂^{ant} is estimated, from Eq. 13, to be $3.7 \mu\text{mol kg}^{-1}$. This represents a 2σ of 7.4 for TCO₂^{ant} which is in excellent agreement with the observed residuals (Fig. 14c) of Eq. 9.

7. Summary

The accurate measurements of 2367 TCO₂ and TA seawater samples from the Gulf of Aden along the nominal 9N latitude across the Arabian Sea and along the nominal 10N latitude across Bay of Bengal illustrate the large spatial variations of biogeochemical properties. At the time of these observations, these variations reached up to $50 \mu\text{mol kg}^{-1}$ and $40 \mu\text{mol kg}^{-1}$ in TCO₂ and TA, respectively. The low total alkalinity concentrations in surface water are associated with the low-salinity waters in the Bay of Bengal. The highest

TCO₂ concentrations in surface water are observed close to the coast in upwelling areas. At depths each water mass is characterized by different TCO₂ and TA concentrations. The data show the TCO₂ signature of the deep boundary current that flows through the Owen Fractures Zone at depths below 3500 m.

In contrast to the precedent modification (Gruber *et al.*, 1996) of the original approach (Brewer, 1978) to the estimation of anthropogenic CO₂ in the ocean, we presented here a different approach based upon the quantitative analysis of water-source mixing. The results of a multiparametric analysis applied to the conservative tracers, temperature and salinity, and nonconservative tracers, oxygen and TA, quantify the relative contributions of each water-source to the observed concentrations. For instance, this analysis indicates that the intermediate waters in the Gulf of Aden is constituted by more than 30% of the Red Sea intermediate waters (RSW). This analysis further quantifies the contrasts between the composition of seawater in the Arabian Sea and in the Bay of Bengal.

The results were then used in an innovative approach with the observed TCO₂ concentrations to quantify the anthropogenic CO₂ in each of the water-sources. There is no hypothesis for the determination of ΔO_2 and ΔTA or for the pre-industrial pCO₂ levels in the atmosphere and in seawater. In addition to the initial choice of the water-sources well constrained by hydrological properties, this quantification of anthropogenic CO₂ depends upon the hypothesis that the coefficients 0.78 and 0.5 of Eq. 5 are valid and constant with time and space. These coefficients have been determined long ago and are based upon the fundamental natural biogeochemical transformation in the ocean that was confirmed by observations (Redfield, 1934; Anderson and Sarmiento, 1994). The uncertainty associated with these coefficients is relatively small. The residuals of the calculation (Eq. 9) used to quantify the concentrations of anthropogenic CO₂ in each of the water-sources approach the standard deviation of the TCO₂ measurements.

The results indicate that in 1995 the surface water-sources contained over 42 $\mu\text{mol kg}^{-1}$ of anthropogenic CO₂. Thus, the anthropogenic signal in the ocean is now clear and can be accurately quantified. As the anthropogenic signal increases over the years, and the frequency and accuracy of the measurements further improves, the relative uncertainty associated with the estimates of the anthropogenic CO₂ concentration in each water-source will decrease.

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