Increase of total alkalinity due to shoaling of aragonite saturation horizon in the Pacific and Indian Oceans: Influence of anthropogenic carbon inputs

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[1] Aragonite saturation horizon (ASH) shallowed significantly by 25 to 155 m and 16 to 124 m in the Pacific and Indian Ocean respectively in two decades. Apparent oxygen utilization (AOU) increased by 3 to 34 and 0.5 to 31.5 μmol kg^{-1} in the Pacific and Indian Ocean respectively at the depth of ASH during this period. DIC increased by 12.5 to 36.8 and 5.5 to 32 μmol kg^{-1} in the vicinity of ASH in the Pacific and Indian Ocean respectively due to combined effect of increased anthropogenic CO_2 and change in AOU. TA increased significantly by 5 to 10 and 4 to 9.2 μmol kg^{-1} in the Pacific and Indian Oceans respectively at the ASH most likely as a result of aragonite dissolution. The upward migration of ASH solely due to anthropogenic CO_2 amounted to 6 to 58 m in the Pacific and 4 to 44 m in the Indian Ocean.   INDEX TERMS: 1615 Global Change: Biogeochemical processes (4805); 4215 Oceanography: General: Climate and interannual variability (3309); 4805 Oceanography: Biological and Chemical: Biogeochemical cycles (1615); 4806 Oceanography: Biological and Chemical: Carbon cycling. Citation: Sarma, V. V. S. S., T. Ono, and T. Saino, Increase of total alkalinity due to shoaling of aragonite saturation horizon in the Pacific and Indian Oceans: Influence of anthropogenic carbon inputs, Geophys. Res. Lett., 29(20), 1971, doi:10.1029/2002GL015135, 2002.

1. Introduction

[2] The carbonate pump is driven by the formation of calcareous skeletons by marine planktonic organisms in the euphotic zone and their subsequent sinking/dissolution to the deep-ocean and underlying sediments [Milliman, 1993]. In recent years, there is concern about the progressive increase in the levels of fossil fuel-derived CO_2 in the atmosphere at a superceding rate of absorption by the oceanic waters [Bates, 2001; Dore et al., 2000]. The current Inter-governmental Panel on Climate Change (IPCC) estimates for the oceanic sink of anthropogenic CO_2 to be 2.0 ± 0.8 Pg C y^{-1} [Prentice et al., 2001]. Recent estimates showed that the highest anthropogenic CO_2 inventories were found in the Pacific [45 ± 15 PgC; Sabine et al., 2001] followed by Atlantic [40 ± 9 PgC; Gruber, 1998] and

Indian Ocean [20 ± 3 PgC; Sabine et al., 1999] with deeper penetration in the Atlantic followed by Pacific and Indian Ocean. The increase in anthropogenic CO_2 inventory since GEOSECS was found to be 4.1 ± 1 PgC for north of 35°S in the Indian Ocean [Sabine et al., 1999]. The present rise in atmospheric CO_2 levels results in decrease in calcification in the surface waters [Riebesell et al., 2000]. Early investigations revealed upward translation of aragonite saturation horizon by 50 to 70 m's in the North Pacific and it was attributed to increase in anthropogenic CO_2 [Feely and Chen, 1982; Feely et al., 1984; Feely et al., 2002]. The dissolution of $CaCO_3$ neutralizes anthropogenic CO_2 and adds TA via the dissolution reaction: $CO_2 + CaCO_3 + H_2O \rightarrow 2HCO_3^- + Ca^{2+}$. The increase in TA serves to enhance the seawater's capacity of CO_2 absorption from the atmosphere. Several investigators observed increase in DIC in the upper layers [Peng et al., 1998; Ono et al., 2000], however, no reports have been made so far on changes in TA at the vicinity of aragonite saturation. In addition to this, the apparent oxygen utilization (AOU) was also found to be increasing with time at least in the past 3 decades in the subsurface waters at the rate of 0.9 μmol kg^{-1} y^{-1} in the North Pacific [Ono et al., 2001]. These changes in DIC and AOU increase the DIC/TA ratio and, consequently, could increase the rate of $CaCO_3$ dissolution below the aragonite and calcite saturation horizons. The goals of this research are to: 1) examine the influence of enhanced anthropogenic CO_2 levels on dissolution of aragonite, 2) changes in TA at the vicinity of aragonite saturation horizon (ASH) and 3) influence of change in AOU on the depth of ASH in about two decades of time scale (1974–1994, i.e., GEOSECS to WOCE) in the World Oceans.

2. Data and Methods

[3] The details of the sample collection and methodologies followed in DIC and TA measurements during WOCE were given elsewhere [Johnson et al., 1998; Millero et al., 1998]. WOCE data are obtained from WOCE Hydrographic Program Office (http://whpo.ucsd.edu/) whereas GEOSECS data downloaded from http://ingrid.ldgo.columbia.edu/SOURCES/.GEOSECS/. The accuracy of the DIC and TA measurements during WOCE was estimated to be ± 3.0 and ± 5 μmol kg^{-1} respectively [Lamb et al., 2002] whereas no such estimations were done during GEOSECS. For each GEOSECS station, we have selected WOCE station which had the same water-mass structure within the range of $\pm 4°$ latitude and $\pm 6°$ longitudes. The salinity difference by 0.02–0.04 units at the potential density <26.5 and ≤ 0.02 at density >26.5 and 27.5 is accepted during selection of stations. Since

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Table 1. The Change in TA, DIC, AOU and ASH Between GEOSECS and WOCE

Stn No.	Lat deg	Long deg	Density σ_0	ΔTA μM	ΔDIC μM	ΔAOU μM	C_{anth} μM	ΔASH[a] (m)	ΔASH[b] C_{anth} (m)
217	44.7 N	176.8 W	26.80	12.6	21.0	5.2	10.7	65 ± 14	6 ± 14
219	53.1 N	177.3 W	26.75	10.8	16.5	5.8	6.7	67 ± 16	6 ± 16
213	30.9 N	168.5 W	26.90	8.0	13.5	5.0	5.7	62 ± 35	7 ± 35
241	4.7 N	179.0 E	26.98	10.2	23.0	14.0	7.2	155 ± 97	18 ± 97
251	4.6 S	179.0 E	27.00	8.0	36.8	34.0	6.7	169 ± 69	50 ± 69
263	16.8 S	167.1 W	27.30	8.0	16.5	3.0	10.2	87 ± 62	58 ± 62
265	17.8 S	165.0 W	27.40	12.0	16.2	4.0	7.1	81 ± 138	56 ± 138
267	19.3 S	171.5 W	27.30	6.0	15.2	4.5	8.7	57 ± 103	46 ± 103
290	58.0 S	174.0 W	27.30	5.5	12.5	7.5	4.0	15 ± 220	29 ± 220
293	52.6 S	178.0 W	27.30	8.8	16.5	5.0	8.3	25 ± 121	15 ± 121
445	8.5 N	86.0 E	26.80	5.0	7.1	1.9	3.1	102 ± 152	32 ± 152
446	12.6 N	84.6 E	26.82	8.3	23.0	7.5	13.1	49 ± 24	19 ± 24
447	5.1 N	80.0 E	26.90	9.2	32.0	31.5	3.2	124 ± 59	9 ± 59
440	9.5 S	95.1 E	27.20	4.0	10.8	6.2	4.1	79 ± 55	27 ± 55
441	5.1 S	91.8 E	27.15	4.2	5.5	0.5	3.0	22 ± 49	4 ± 49
449	5.0 S	80.1 E	27.10	4.2	8.2	1.0	5.4	112 ± 92	44 ± 92
451	15.0 S	80.0 E	27.30	5.5	12.8	8.5	3.5	16 ± 115	32 ± 115

The errors associated with ΔTA and ΔDIC are ±7 and 5.8 μmol kg^{-1} respectively.
$\Delta-$Denotes difference between WOCE to GEOSECS except ΔASH which is GEOSECS-WOCE.
[a]Difference in depth of ASH between GEOSECS to WOCE.
[b]Difference in depth of ASH between GEOSECS to WOCE driven by anthropogenic carbon.

there were no CRM standards available during GEOSECS time, in order to maintain consistency of both data sets, GEOSECS data have been corrected with reference to WOCE assuming deep waters are not changed since last two decades. Since intermediate waters are found to be changed due to climatic variations [for instance, *Watanabe et al.*, 2001], we decided to correct GEOSECS data set with reference to deep-water data. In order to perform this, all the data were linearly interpolated to σ_θ = 24.80 to 27.75. Deep water data (depth >3000 m) of GEOSECS were linearly fit to the WOCE vertical profile and appropriate corrections were made to the entire profile assuming the offset between two stations is constant with depth. The uncertainties involved in these corrections are ±2–3 μmol kg^{-1}. The total errors associated with the estimation of ΔDIC and ΔTA is ±5.8 and ±7.0 μmol kg^{-1} respectively.

[4] The CO_3^{2-} ion concentrations are derived from TA and TC pair using *Mehrbach et al.* [1973] dissociation constants that have been refitted by *Dickson and Millero* [1987]. The solubility product is calculated for aragonite based on *Mucci* [1983].

3. Results and Discussion

[5] North-south variability in the degree of aragonite saturation in the central Pacific (WOCE leg 15), Indian (Leg I09) and western Atlantic (Legs A17, A22) Oceans is depicted in Figures 1a–1c. The shallowest ASH is found at ∼120 m in the northern Pacific (north of 40°N) whereas it is deepened to ∼1400 m in the South Pacific. Such shallow ASH was observed earlier in the northwest and central Pacific [*Feely et al.*, 1984]. On the other hand, the deepest ASH is found in the western Atlantic and the trend is completely opposite to that of Pacific (Figure 1b). The ASH is deeper (∼2900 m) in the north (40°N) and it was shoaled to ∼1000 m at 47°S (Figure 1b). The deeper saturation depth in the north could be due to formation of fresh deep water masses within the Norwegian and Greenland Seas that results in deepening of the ASH. The shallow ASH farther south could be related to upward migration of these water masses in the

south of 40°S [*Reid and Lyan*, 1971]. Indian Ocean showed a similar kind of distribution pattern as in the Pacific, with the deepest ASH (∼1200 m) near 30°S and shallowest (240 m) in the north (19.7°N) (Figure 1c). Such shallow ASH in the Bay of Bengal was attributed to intense regeneration of organic matter [*George et al.*, 1994].

[6] Since pre-industrialization era, anthropogenic carbon dioxide inputs have caused atmospheric CO_2 levels to increase from 280 to 370 ppmv [*Keeling and Whorf*, 2000]. The increase in DIC in the surface layers was found to be 0.61 to 1.85 μmol kg^{-1} y^{-1} at BATS [*Bates*, 2001] and 0.79 to 1.58 μmol kg^{-1} y^{-1} at HOT [*Dore et al.*, 2000] stations, which seems to be in pace with atmospheric pCO_2. Recently *Feely et al.* [2002] found upward translation of ASH by 30–100 m in the North Pacific and 30–80 m in the South Pacific since pre-industrial times. This estimate is an average rate of ASH shallowing since pre-industrial era to the present. The anthropogenic carbon inputs to the atmosphere were changed significantly with time and it is found to be much higher during recent decades [*Keeling and Whorf*, 2000]. In order to examine the change in depth of ASH due to increased anthropogenic CO_2 inputs and consequent influence on TA in recent years in the World Oceans, we compared the data collected during GEOSECS with that of recently collected WOCE at selected locations.

[7] The vertical structure of aragonite saturation-state in the Pacific and Indian Oceans suggest that it is shallowed substantially compared to GEOSECS time (Table 1). For instance, the transition of super-saturation to under-saturation with respect to aragonite occurred in the North Pacific at potential density surface of 26.940 (depth of 740 ± 33 m GEOSECS st. 213) during 1974, whereas this transition was occurred at potential density of 26.822 (678 ± 13 m) during 1994 (WOCE) almost at the same location. It was observed that anthropogenic carbon penetrates up to a depth of ∼1500 m in the Pacific and the ASH is within the depth of penetration where >25 μmol kg^{-1} of anthropogenic carbon accumulated since pre-industrial times [*Sabine et al.*, 2001]. The deep ASH, found at 27.841 (2852 ± 141 m; GEOSECS st. 48) potential density surface in the South
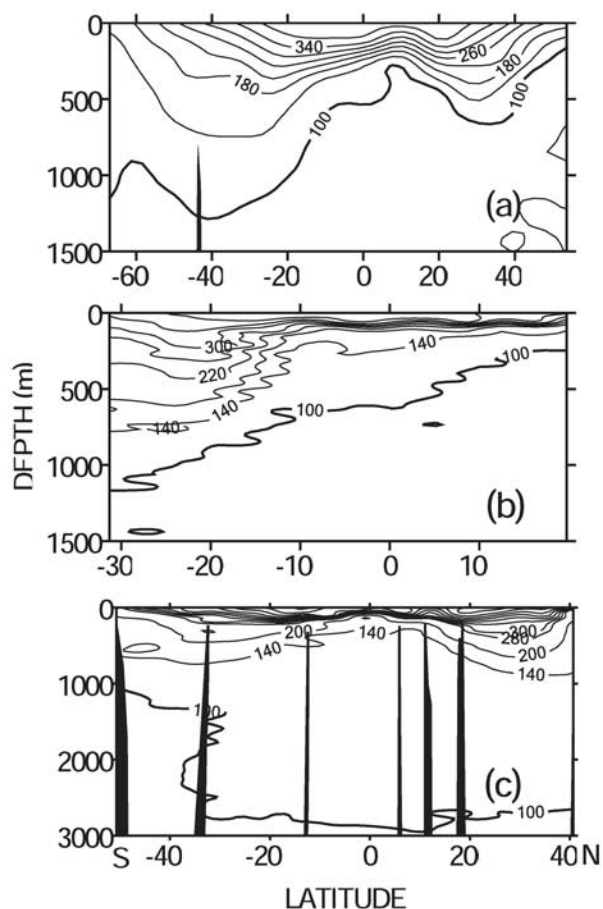
Figure 1. Distribution of aragonite saturation (omega, %) in the (a) central Pacific Ocean (WOCE leg P15), (b) Indian Ocean (WOCE leg I09) and (c) Western Atlantic Ocean (WOCE legs A17 and A22). The waters above 100% contour are super-saturated and below are under-saturated with respect to aragonite.

Atlantic Ocean during GEOSECS [1973] is found at 27.852 (2783 ± 173 m) potential density during WOCE observation [1994]. These differences in the Atlantic are within the errors involved in the measurements of DIC and TA of GEOSECS. *Gruber* [1998] observed that anthropogenic CO_2 penetrates deeper even up to the bottom ($\sim$5 μmol kg^{-1}) since pre-industrial times, however, it is quite small to detect significant changes in the depth of ASH with less precise GEOSECS data. On the other hand, ASH occurred at potential density of 26.882 (385 ± 16 m; GEOSECS st. 446) during GEOSECS [1978], that has been migrated to 26.765 density surface (336 ± 18 m) during WOCE [1995] in the northern Indian Ocean. This feature is consistent with depth of anthropogenic carbon penetration in the Indian Ocean [*Sabine et al.*, 1999].

[8] It is unlikely that such large changes in depth of ASH were brought only by the accumulation of anthropogenic CO_2 itself since GEOSECS time. For instance, if we assume that DIC has increased under equilibrium conditions with respect to atmospheric pCO_2 increase [from 330 to 355 ppm over the period 1973–1994; *Keeling and Whorf*, 2000], the expected increase in DIC would be 17–19 μmol kg^{-1}. On contrary to this, the observed increase in DIC is much higher

than expected (Table 1). The increase in DIC (ΔDIC) is ranged from 12.5 to 36.8 μmol kg^{-1} in the Pacific and 7.1 to 32 μmol kg^{-1} in the Indian Ocean. No significant changes in DIC are found in the Atlantic (not shown in Table 1). The alternative possibility to bring such large changes in DIC could be due to change in either organic matter decomposition rates or residence time of waters since last two decades. In order to examine such possibilities, we compared the AOU levels between GEOSECS and WOCE at the stations studied here (Table 1). The AOU is found to be increased at the depth of ASH during WOCE compared to GEOSECS in the World Oceans. Comparatively higher AOU changes were observed in the Pacific (4 and 34 μmol kg^{-1}) than Indian Ocean (0.5–8.5 μmol kg^{-1} except at sta. 447) at the depth of ASH. The increase in AOU due to enhanced organic matter decomposition rates could be possible as *Emerson et al.* [2001] suggested that biological pump has increased as a result of onset of nitrogen fixation in the recent several decades. On the other hand, the observed AOU increase was attributed to change in the residence time of each water mass by *Watanabe et al.* [2001] who found the increase in CFC-age of subsurface water masses in the North Pacific. This could be possible as CFC age of water mass can not be influenced by any biological processes. Moreover, *Ono et al.* [2001] observed temporal increase in AOU by 0.9 μmol kg^{-1} y^{-1} in the subsurface waters of the western sub-arctic Pacific from 1968 to 1998 and attributed it to decrease in vertical water exchange. Such increase in AOU would significantly enhance DIC levels and results in change in DIC/TA ratio that ultimately governs depth of ASH. Assuming the C/O ratio of 0.768 [*Redfield et al.*, 1963], we can estimate that about 3 to 26 μmol kg^{-1} of DIC is increased due to change in AOU at the depth of ASH in the Pacific and $\sim$0.4 to 24.6 μmol kg^{-1} in the Indian Ocean (Table 1). Interestingly, TA is also found to be increased by 5.5–12.6 and 4.0 to 12 μmol kg^{-1} in the Pacific and Indian Ocean, respectively (Figure 2 and Table 1) only in the vicinity of

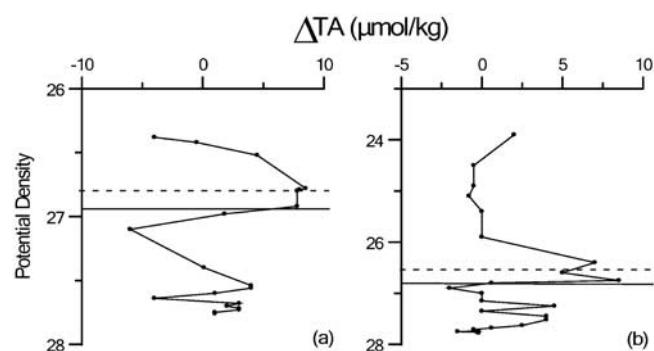**Figure 2.** The change in total alkalinity (μmol/kg) with reference to potential density (σ_0), between GEOSECS and WOCE in the (a) North Pacific [GEOSECS #213, 30.97°N, 168.48°W and WOCE leg P15NA, #50, 31.00°N, 165.00°W (b) North Indian Ocean [GEOSECS #446, 12.6°N, 84.6°E and WOCE leg I09, #241, 13.86°N, 91.5°E]. Horizontal lines show aragonite saturation during GEOSECS (solid line) and WOCE (dashed line). Increase in TA between two horizontal lines represents the influence of dissolution of aragonite skeletal material due to increased anthropogenic carbon inputs during past two decades.

ASH. These results suggest that the TA has increased because of aragonite dissolution over the past two decades. Comparatively, this increase is more significant in the northern basins of Pacific and Indian Ocean than south. On the other hand, no statistically significant increase is found in the Atlantic Ocean. Recently *Feely et al.* [2002] observed that the *in situ* $CaCO_3$ dissolution rates are increased sharply to $\sim 0.35 \, \mu mol \, kg^{-1} \, y^{-1}$ in the vicinity of 26.8 potential density surfaces in the North Pacific. If this dissolution rate is increased due to upward migration of ASH around this density surface, this would increase TA by 7 $\mu mol \, kg^{-1}$ in two decades of time scale (1973 to 1994). This is in good agreement with our observation of increase in TA by an average of $\sim 9 \, \mu mol \, kg^{-1}$ in the Pacific. Moreover, the dissolution of aragonite not only enhances TA but also DIC by half of TA increase. After correcting for this as well as for AOU change, the increase in DIC due to anthropogenic CO_2 at the depth of ASH is estimated to 4 to 10.7 $\mu mol \, kg^{-1}$ in the Pacific (average 7.3 $\mu mol \, kg^{-1}$), 3 to 13.1 $\mu mol \, kg^{-1}$ (average 8 $\mu mol \, kg^{-1}$) in the Indian Ocean respectively since 1973. These results are in good agreement with observations of *Ono et al.* [2000] who found increase in DIC by 5–10 $\mu mol \, kg^{-1}$ since 1973 to 1993 in the vicinity of 26.8 potential density surfaces in the north western Pacific due to injection of anthropogenic CO_2. The upward migration of depth of ASH, due to accumulation of anthropogenic CO_2 during study period, is observed to be about 6 to 58 m in the Pacific, which is about 6–80% to that of total migration of ASH, whereas 4 to 32 m (7 to 40%) in the Indian Ocean. Nevertheless, increase in TA at the vicinity of depth of ASH is more important as this enhances ocean's capacity to absorb more anthropogenic CO_2. Though this analysis results have large error bars on shallowing of ASH by anthropogenic carbon (Table 1, column 10), due to less precise GEOSECS inorganic carbon data set, however, this kind of analysis should be performed, in future, with long time series highly precise data at several regions in the World Oceans in order to understand oceanic response on continuously increasing anthropogenic carbon inputs.

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