

Decadal-scale increases of anthropogenic CO₂ in Antarctic Bottom Water of the Indian sector of the Southern Ocean

*Akihiko Murata¹, Yuichiro Kumamoto¹, Kenichi Sasaki¹

1. Japan Agency for Marine-Earth Science and Technology

The Southern Ocean is regarded as one of the most important sinks for global carbon budget, because it is estimated that approx. 40% of ocean uptake of anthropogenic CO₂ (hereafter abbreviated as antCO₂) occurs in the Southern Ocean. In the ocean, it is known that Sub-Antarctic Mode Water and Antarctic Intermediate Water, both of which are Southern Ocean-origin, play a large role in absorbing antCO₂ and transporting it northward. However, for role of Antarctic Bottom Water (AABW) in accumulating and absorbing antCO₂, it is still an open question. In the 1980's, it was said that that accumulation and uptake of antCO₂ in AABW is small (Chen, 1982; Poisson and Chen, 1988), because existence of sea ices effectively blocks air-sea exchange of CO₂, and because signals of antCO₂ are diluted by mixing with water (e.g., Circumpolar Deep Water) of no or little contamination of antCO₂. Recent studies reveal that AABW is also contaminated significantly by antCO₂, although it is not so high (Rios et al., 2012; Pardo et al., 2014). Furthermore, influences of ocean climate changes such as warming, desalination, acidification, etc. on ocean uptake of antCO₂ remain unclear.

To elucidate how large AABW, defined as neutral density of $\sigma_\theta \geq 28.27 \text{ kg m}^{-3}$, takes up antCO₂, we examined decadal-scale increases of antCO₂ (ΔantCO_2) along a zonal section at nominal 62°S ranging from 30°E to 160°E in the Indian sector of the Southern Ocean. For the purpose, we used high-quality data for CO₂-system and related properties collected about 17 years apart in 1994/1995 and 2012/2013. These data were obtained under international observation programs such as World Ocean Circulation Experiment and Global Ship-based Hydrographic Investigations Program. From depth - longitude section of ΔantCO_2 , it was found that there was a clear contrast of distributions of ΔantCO_2 in AABW between the eastern and western sides of the Kerguelen Plateau. That is, higher increases of $> 5 \text{ mmol kg}^{-1}$ were found in the eastern side, while lower increases or even decreases were found in the western side. In the eastern side, in contrast to previous studies, increases of antCO₂ were largest ($> 9.0 \text{ mmol kg}^{-1}$) in the bottom water, i.e., AABW. The higher increases were especially conspicuous east of 110°E. Significant increases of antCO₂ in bottom and deep waters were detected through the section, although they became gradually reduced in magnitude and depth range westward from 110°E. Vertical distributions of ΔantCO_2 showed significant positive correlations with decadal-scale changes in CFC-12 and with distributions of SF₆, both of which can be used as a proxy of ocean circulation and ventilation, meaning that the distributions were mainly controlled by physical processes. Comparison of ΔantCO_2 between calculation methods with and without total alkalinity presented differences of ΔantCO_2 west of 50°E (the latter was smaller than the former). This may be related to decreases in production of particulate inorganic carbons in the Southern Ocean (Freeman and Lovenduski, 2015). The highest storage rate of antCO₂ was estimated to be $1.1 \pm 0.6 \text{ mol m}^{-2} \text{ a}^{-1}$ at longitudes 130°–160°E, which is a value integrated from surface to bottom layers of statistically-significant ΔantCO_2 . With this condition of integration, we believe that the value is conservative. West of the Kerguelen Plateau, the storage rate was $0.2 \pm 0.1 \text{ mol m}^{-2} \text{ a}^{-1}$ at most. The contrast is due probably to differences of formation regions of AABW; west of 80°E (Kerguelen Plateau), the AABW consists mostly of Weddell Sea-origin water, while east of it, the AABW consists of both the Adélie coast- and Ross Sea-origin waters.

The above results highlight that processes for uptake and accumulation of antCO₂ by the bottom water work well in the Indian sector of the Southern Ocean, at least, in the eastern part of it.

Keywords: Anthropogenic CO₂, Antarctic Bottom Water (AABW), Southern Ocean

Underway measurements of surface $p\text{CO}_2$ and total alkalinity in Kuroshio-Oyashio transition region

*Naohiro Kosugi¹, Daisuke Sasano¹, Masao Ishii¹, Eitarou Oka², Chiho Sukigara³

1. Meteorological Research Institute, 2. Atmosphere and Ocean Research Institute, The University of Tokyo, 3. Nagoya University

We made measurements of surface partial pressure of CO_2 ($p\text{CO}_2$) and total alkalinity (TA) in Kuroshio-Oyashio transition region off the eastern Japan in June 2016. Surface TA was measured every 15 minutes. Spatial resolution of TA was 6 km in case of cruising at 12 knot.

Surface TA (open circle in Figure 1) showed fine spatial variation which can not be captured by the interpolation of the bottle sampling and measurement of TA (open square in Figure 1) at CTD station which located every 2 degrees in longitude. The estimation of TA by Lee *et al.* [2006] (gray dot in Figure 1) overestimated the measurement by up to 30 $\mu\text{mol/kg}$.

We calculated surface DIC from TA and $p\text{CO}_2$ obtained by the underway measurements. Calculated DIC was in good agreement with the measured DIC taken from nearby CTD station. Difference and standard deviation between calculated and measured value were 0.8 and 5.4 $\mu\text{mol/kg}$ respectively ($N = 38$).

Underway measurement of $p\text{CO}_2$ and TA can reproduce other carbonate parameters such as DIC and pH accurately. This method is beneficial to understand carbon cycling in coastal region and Kuroshio-Oyashio transition region where spatial variation of TA is large.

The variation of TA by precipitation and evaporation can be excluded by salinity normalization.

Normalized alkalinity to Salinity = 35 (NTA_{35}) has a large meridional gradient in the western North Pacific. NTA_{35} in the subtropical region and subarctic gyre were about 2300 and 2370 $\mu\text{mol/kg}$ respectively [Takatani *et al.* 2014]. In our observation, NTA_{35} also showed large zonal variation and ranged 2310-2355 $\mu\text{mol/kg}$ along 41°N and 2300-2325 $\mu\text{mol/kg}$ along 37.5°N. These dynamic spatial variation of NTA_{35} was attributable to the complicated distribution of Kuroshio and Oyashio water. $p\text{CO}_2$ was concave against NTA_{35} and the smallest around $\text{NTA}_{35} = 2320 \mu\text{mol/kg}$. Below this NTA_{35} , $p\text{CO}_2$ increased thermodynamically due to temperature rise. On the other hand, nutrients was significantly high above this NTA_{35} . This indicated that $p\text{CO}_2$ was high because the DIC supplied to the surface by the winter mixing had not been substantially reduced by biological production.

Keywords: Kuroshio-Oyashio transition region, Total alkalinity, $p\text{CO}_2$

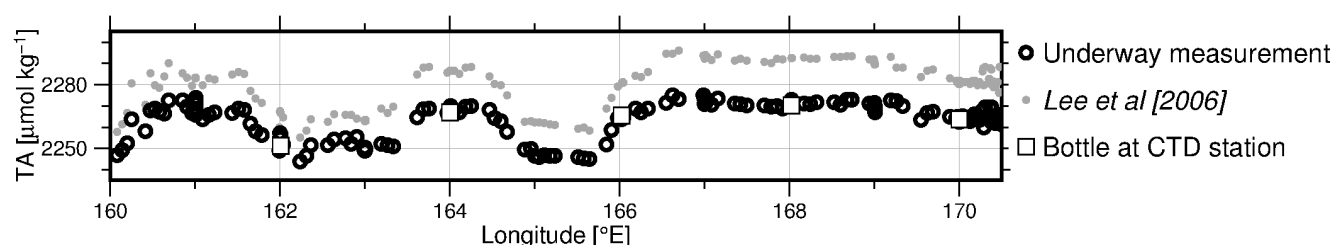


Figure 1 Zonal variation of surface total alkalinity along 41°N in June 2016.

Estimation of the increase in the anthropogenic carbon dioxide column inventory in the 137°E section by the eMLR method

*Shinji Masuda¹, Daisuke Sasano², Kazutaka Enyo¹, Yosuke Iida¹, Atsushi Kojima¹, Masao Ishii², Toshiya Nakano¹

1. Japan Meteorological Agency, 2. Meteorological Research Institute

The ocean absorbs approximately 30% of the carbon dioxide (CO₂) released into the atmosphere by human activities such as fossil fuel combustion and land-use changes, and is mitigating the progress of global warming. The western North Pacific subtropical zone is considered as one of the strong sinks of the anthropogenic CO₂ in the world oceans. It is important to evaluate the change in its column inventory in this area for the better understanding the global carbon cycle change, thereby better projection of the global warming. Japan Meteorological Agency has been carried out the atmospheric and oceanic CO₂ measurement along its repeat line at 137°E every year since early 1980s. The WOCE/GO-SHIP's precise measurements with high-resolution and full water-column samplings have been made in the years 1994, 2010 and 2016. In this study, we evaluated the changes in the column inventory of the anthropogenic CO₂ in the 137°E section based on the biogeochemical data collected in these cruises using the eMLR method (Friis et al. 2005). In our eMLR analysis, we used the relationship between salinity-normalized preformed DIC (nDIC*) and chemical tracer NO (Broecker, 1974).

$$\text{nDIC}^* = \{\text{DIC} - 117/170 \cdot (\text{O}_2^{\text{sat}} - \text{O}_2)\} \cdot 35/S,$$

$$\text{NO} = \text{O}_2 + 170/16 \cdot \text{NO}_3^- \quad (\text{Anderson and Sarmiento, 1994}).$$

NO is preformed oxygen concentration calculated using nitrate concentration. It is a quasi-conservative tracer that does not change with biological activity in the same water mass.

nDIC* and NO are linearly correlated to each other below the winter mixed layer in the subtropical zone between 19°N and 34°N:

$$\text{nDIC}^* = a + b \cdot \text{NO}.$$

In the tropics to the south of 18°N, their relationships show non-linearity due to the north-south fluctuation of the North Equatorial Current and mixing with the water mass from the Southern Ocean. We then calculated the differences in nDIC* among three cruises in 1994, 2010 and 2016 in the subtropics between 19°N and 34°N using the difference in the slope *b* in the above equation, and integrated over the water column above $\sigma_\theta = 27.3$ to estimate the column inventory of anthropogenic CO₂. The rates of increase in the anthropogenic CO₂ inventory were 0.83, 0.67 and 0.62 mol/m²/year (preliminary) at 30°N, 25°N and 20°N, respectively, for 22 years between 1994 and 2016, and 1.84, 1.57 and 1.44 mol/m²/year (preliminary) at 30°N, 25°N and 20°N, respectively, for 6 years between 2010 and 2016 (Fig. 1). The higher rate in the northern subtropics is attributable to the deeper ventilation (Fig. 2). It is also higher for the period from 2010 to 2016 than that from 1994 to 2016. We will also study in the different water masses such as those in the tropical zone and above the bottom of winter mixed layer where the relationship between nDIC* and NO shows non-linearity.

Keywords: anthropogenic CO₂, 137°E section, column inventory

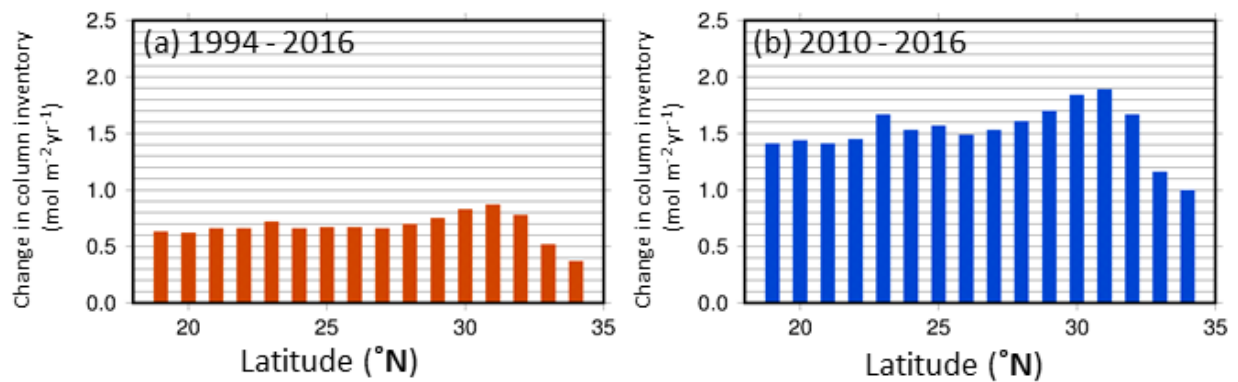


Fig. 1 Column inventory rates of anthropogenic CO₂ along 137E between (a) 1994 and 2016 and (b) 2010 and 2016.

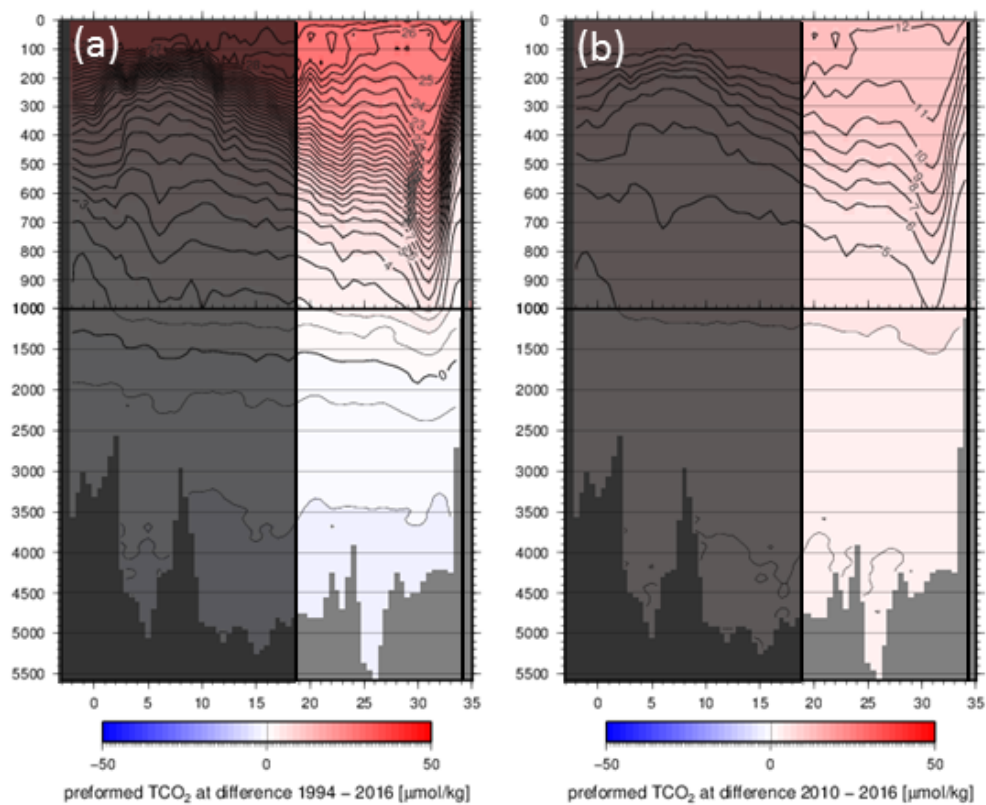


Fig. 2 Sections of anthropogenic CO₂ changes between (a) 1994 and 2016 and (b) 2010 and 2016. Gray colorings indicate areas outside the subtropical gyre.

Lead spreading through sub-surface water over the North Pacific

*Yoshiki Sohrin¹, Linjie Zheng¹, Tomoharu Minami¹, Shotaro Takano¹

1. Institute for Chemical Research, Kyoto University

The international research programme GEOTRACES is revealing global distributions of trace elements in the ocean (<http://www.geotraces.org>). We have been studying the distributions of Al, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb in seawater using our chelating resin extraction-ICP-MS method (Minami et al., 2015) as a part of Japan GEOTRACES. This presentation reports our novel results from the North Pacific.

Seawater samples were collected during R/V Hakuho Maru cruises of KH-05-2 (along 160°W), KH-11-7 (along 165°E), and KH-12-4 (along 47°N) using a clean sampling system. The vertical profiles of dissolved Al, Mn, Co, Ni, Cu, and Cd were quite similar between cross-over stations of KH-05-2 and KH-12-4.

However, a sub-surface maximum of dissolved Pb significantly decreased from 2005 to 2012. The decrease is consistent with the decrease of Pb in sub-surface water in the North Atlantic (Lee et al., 2011). On the ocean section along 160°W, the sub-surface Pb maximum was observed to the north of 10°N with a core at ~35°N and ~200 m depth. On an isopycnic surface of $\sigma_0 = 26.00$, the sub-surface Pb maximum decreased from 165°E to 130°W. In addition, both the dissolved Pb and Co showed scavenged profiles with a strong correlation >300 m depth for all stations. Although the concentrations of Co were high in surface waters near continents, those of Pb were high in surface waters beneath westerlies.

It is widely recognized that the sub-surface Pb maximum is caused by anthropogenic sources and the decrease in the Atlantic is due to the phase-out of leaded gasoline in the United States and Europe (Lee et al., 2011). Our data suggest that the decrease is slower in the Pacific. It is very likely that anthropogenic lead is majorly caused by coal burning in China, carried to the North Pacific by westerlies, dissolved in mixing layers, and spreaded through sub-surface water over the North Pacific by current systems.

Lee, J.-M., Boyle, E.A., Echegoyen-Sanz, Y., Fitzsimmons, J.N., Zhang, R. and Kayser, R.A. (2011) Analysis of trace metals (Cu, Cd, Pb, and Fe) in seawater using single batch nitrilotriacetate resin extraction and isotope dilution inductively coupled plasma mass spectrometry. *Anal. Chim. Acta* 686, 93-101.

Minami, T., Konagaya, W., Zheng, L., Takano, S., Sasaki, M., Murata, R., Nakaguchi, Y. and Sohrin, Y. (2015) An off-line automated preconcentration system with ethylenediaminetriacetate chelating resin for the determination of trace metals in seawater by high-resolution inductively coupled plasma mass spectrometry. *Anal. Chim. Acta* 854, 183-190.

Keywords: lead, GEOTRACES, North Pacific

Speciation of inorganic iodine in bottom water of the Funka Bay, Hokkaido

*Atsushi Ooki¹, Misato Kyoya¹, Ayane Mori¹, Daiki Nomura¹, Hiroji Onishi¹, Kenshi Kuma¹, Hajime Obata²

1. Hokkaido University, 2. The University of Tokyo

Dissolved iodine is typically present as inorganic compounds, iodate (IO_3^-) and iodide (I^-), in seawater at concentrations approximately 500 nmol/L. In oxic seawater, IO_3^- (400 - 500 nmol/L) is more stable than I^- (< 100 nmol/L). In hypoxic seawater, IO_3^- is reduced to I^- by abiotic chemical reaction. Reduction of IO_3^- in oxic surface seawater is believed to be relevant to nitrate reductase activity in phytoplankton cell and nitrate reducing bacteria (e.g. Tsunogai and Sase, 1969). Recent studies have proposed large variety of abiotic / biological I^- oxidation processes in seawater.

In this study, the objective was to investigate the temporal variation of iodine speciation in high productive coastal environment in subarctic area. Ship observations were conducted in Funka Bay, Hokkaido, in February, March, April, May, July, August, October, December of 2016. Seawater samples were vertically collected by CTD-observation system and Van-Dorn water sampler in the basin area of the Bay. The sampling depths were 5, 10, 15, 20, 25, 30, 40, 50, 60, 70, 75, 80, 85, 90 m. Nutrients ($\text{NO}_3 + \text{NO}_2$, NH_4 , PO_4 , SiO_2) were analyzed by colorimetric method, and I^- was analyzed by voltammetry (CSV). Total inorganic iodine was determined by the CSV analysis of I^- after the ascorbic reduction of IO_3^- (Total I: $\text{T-I} = \text{I}^- + \text{IO}_3^-$).

The total column concentrations of T-I (560 - 580 nmol/L), which were averaged from the surface to the bottom water concentrations, had not distinct maximum or minimum throughout the year. On the other side, in the bottom water (90m), I^- concentrations increased from May (108 nmol/L) to July (150 nmol/L), and IO_3^- concentrations drastically decreased from May (432 nmol/L) to July (188 nmol/L). The N^* value ($= \text{NO}_2 + \text{NO}_3 + \text{NH}_4 - 16 \times \text{PO}_4$), which is used as an indicator of denitrification in seawater, was decreased from April ($-6 \mu\text{mol/L}$) to May ($-12 \mu\text{mol/L}$) and July ($-14 \mu\text{mol/L}$), implying that denitrification occurred in the bottom sediment. We considered that the decrease of IO_3^- concentration from May to July was resulted from the IO_3^- reduction in bottom sediment accompanied by nitrate reduction (denitrification).

Keywords: denitrification, coastal water, redox

Relationship between dissolved zinc and silicate in the western North Pacific from subarctic to subtropical regions

*Taejin Kim¹, Hajime Obata¹, Hiroshi Ogawa¹, Toshitaka Gamo¹

1. The University of Tokyo

Introduction

Zinc (Zn) is an essential trace nutrient for marine microorganisms and one of key parameters in international GEOTRACES project. Recent studies have revealed basin-scale distributions of dissolved Zn and the relationship between dissolved Zn and silicate (Roshan and Wu, 2015; Wyatt et al., 2014). In the subarctic North Pacific, the relationship between dissolved Zn and silicate is very different from that in other open oceans. The decoupling of dissolved Zn and silicate occurs in the subarctic North Pacific intermediate water (sigma-theta: 26.6 –27.5), whereas in both shallow and deep waters generally similar trends to those of the subtropical North Pacific were observed (Kim et al., submitted). In this study, we will present the extensive zonal section full depth transect data of dissolved Zn across the subarctic and subtropical western North Pacific to investigate the influences of the subarctic North Pacific intermediate water by using relationship between dissolved Zn and silicate.

Methods

Seawater samples were collected in the western North Pacific during the R/V Hakuho-maru KH-12-3 cruise (from July to August 2012), by using acid-cleaned Teflon-coated X-Niskin samplers deployed on CTD-CMS. Zn in seawater was determined with cathodic stripping voltammetry (CSV) with ammonium 1-pyrrolidinedithiocarbamate (APDC) after UV-digestion (Kim et al., 2015).

Result and Discussion

The section distributions of dissolved Zn in the western North Pacific are similar to those of silicate. However, the relationships between dissolved Zn and silicate clearly indicate a decoupling of dissolved Zn and silicate in subarctic regions. Based on the relationship between dissolved Zn and silicate in the eastern subtropical North Pacific (Bruland, 1980; Conway and John, 2015), we calculated Zn* as follows.
$$\text{Zn}^* = [\text{Zn}] - 0.052 \times [\text{silicate}] + 0.305$$

High Zn* values were observed in the intermediate water masses from subarctic to subtropical regions. Therefore, Zn* could be a very useful tracer to investigate the influence of the subarctic North Pacific intermediate water on the entire western North Pacific.

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Keywords: Zinc, Trace metal, North Pacific

Potentiality for Over-Estimation of Total Alkalinity observed in Arctic Ocean by Spectrophotometric Method

*Tomonori WATAI¹, Emi DEGUCHI¹, Atsushi ONO¹, Nagisa FUJIKI¹

1. MARINE WORKS JAPAN LTD.

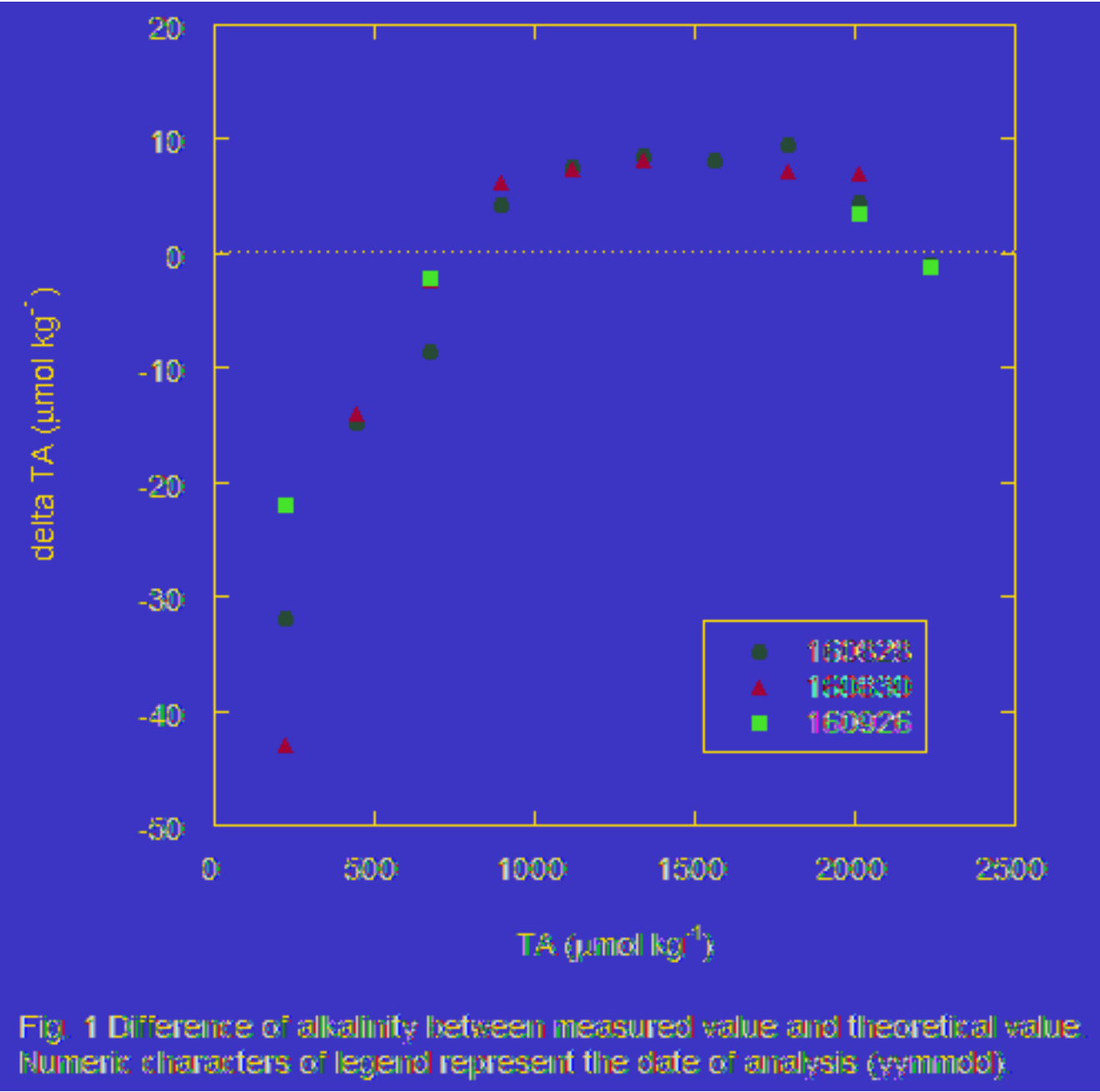
Total alkalinity is an important component of oceanic carbonate system, which deeply affects and is infected to the oceanic chemistry, and also oceanic biology. Measurement of total alkalinity is conducted by potentiometric or spectrophotometric method. Potentiometric method is a traditional method that has been employed for a long time, while spectrophotometric method, that avoids problems attributed to glass-made pH electrodes and has higher precision than potentiometric method, was developed in 1990's and goes into use in this century. A spectrophotometric method was introduced to the laboratory of Research and Development Center for Global Change, Japan Agency for Marine-Earth Science and Technology (JAMSTEC) and the onboard measurement has been conducted for around a decade. Observations were also done in Arctic Ocean, where low alkalinity with its value under $2000 \mu\text{mol kg}^{-1}$ was often observed especially in sea surface. However, spectrophotometric method is suitable for the measurement of oceanic seawater with its alkalinity values ranges from about $2000 \mu\text{mol kg}^{-1}$ to about $2500 \mu\text{mol kg}^{-1}$, and its application for lower alkalinity has not been examined. In this study, we prepared some seawater samples with their alkalinity under $2000 \mu\text{mol kg}^{-1}$, and measured their value by spectrophotometric method.

Reliability of low alkalinity value measured by the spectrophotometric alkalinity system is evaluated by measurements of CRM and its dilutions with ultra pure water under assumption that alkalinity of ultra pure water is $0 \mu\text{mol kg}^{-1}$. Ten types of dilutions with its dilution ratio from 0 % (stock solution) to 90 % are prepared for measurements. Titration should be terminated at $\text{pH} \sim 3.8 - 4.2$ in the case of using BCG as pH indicator. To examine the pH termination ranges are appropriate for analysis of seawater with low alkalinity, the CRM dilutions were analyzed at their pH termination $\sim 3.1 - 4.7$.

Measured alkalinity is not necessarily equal for the designated terminated $\text{pH} \sim 3.8 - 4.2$. The difference of measured alkalinity is in $2 - 3 \mu\text{mol kg}^{-1}$ for the dilution ratio under 40 % (over alkalinity of $1341.9 \mu\text{mol kg}^{-1}$), however, the difference of alkalinity in this pH range increases as the dilution ratio increase (lower alkalinity). The difference becomes about $10 \mu\text{mol kg}^{-1}$ with its dilution ratio of 50 % ($1118.3 \mu\text{mol kg}^{-1}$), and over $20 \mu\text{mol kg}^{-1}$ with its ratio of 80 % ($447.3 \mu\text{mol kg}^{-1}$). Trueness of measured alkalinity is then discussed. Figure 2 shows the difference between the measured alkalinity and theoretical alkalinity. The difference is related to their alkalinity and shows a convex distribution. Around alkalinity of $2250 \mu\text{mol kg}^{-1}$, the averaged alkalinity and theoretical value has good agreement. Below $2250 \mu\text{mol kg}^{-1}$, the difference increases with decrease of alkalinity and measured alkalinity is higher than theoretical alkalinity. Its difference is about $10 \mu\text{mol kg}^{-1}$ around the alkalinity of $1500 \mu\text{mol kg}^{-1}$. Lower than the alkalinity of $1500 \mu\text{mol kg}^{-1}$, the difference decreases as the alkalinity decrease, and the measured value and the theoretical value is nearly equal again at around alkalinity of $750 \mu\text{mol kg}^{-1}$. Lower than the alkalinity of $750 \mu\text{mol kg}^{-1}$, the measured alkalinity turns to be lower than the theoretical alkalinity and its absolute difference increases with alkalinity decrease.

In Arctic Ocean, where sea-ice melting and increase of inflow of river water has a lot of attention in these decades, alkalinity less than $2000 \mu\text{mol kg}^{-1}$ was often observed. On edge of sea-ice melting area, alkalinity less than $1700 \mu\text{mol kg}^{-1}$ is occasionally observed. Our analysis shows that measured alkalinity with its value of around $1300 - 1800 \mu\text{mol kg}^{-1}$ is considered to be overestimated with its difference of around $10 \mu\text{mol kg}^{-1}$. The degree of overestimation is less at external side of this range.

Keywords: Alkalinity, Spectrophotometric Method, Arctic Ocean



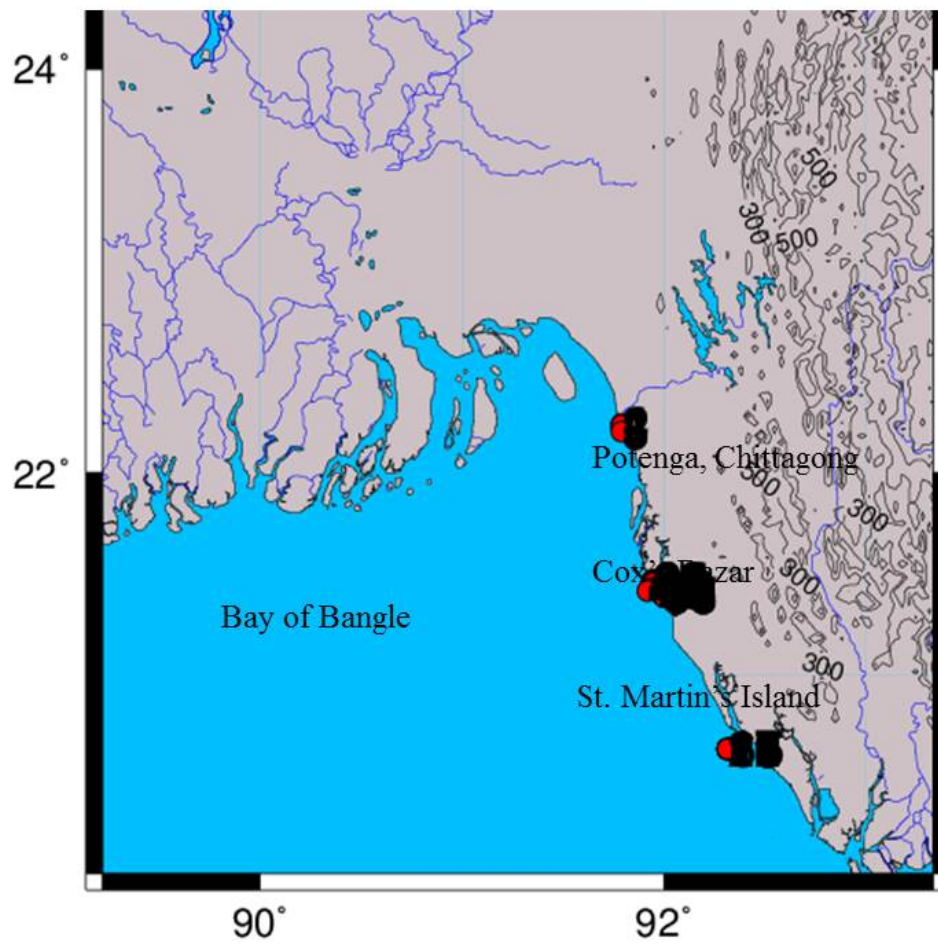
Distribution of Heavy Metals in Beach Water and Sediment of the Bay of Bengal Coast, Bangladesh

*Mst. Shamsun Nahar¹, Md. Nurunnabi Mondal², KOKI Kato³

1. Earth Consultant Co. Ltd, Toyama, Japan, 2. Assistant Professor, Department of Fisheries Management, Bangabandhu Sheikh Mujibur Rahman Agricultural University, Gazipur, Bangladesh, 3. University of Toyama

The objectives of this research are to characterize the distribution of arsenic (As) including other heavy metals in water and sediment, types of coastal sediment in 3 coastal sites, i) located at Cox's Bazar, is the longest unbroken sea beach in the world, running 120 kilometres, ii) Patenga, a sea beach located 14 kilometres south of the port city of Chittagong, iii) St. Martin's Island, a small island (area only 8 km²) from watershed of the Bay of Bangle was investigated. In order to evaluate the occurrence of trace metals, sixty (60) sediment samples, thirty (30) seawater samples, were collected from the three coastal sites. The studied sediments show lower values (2.0-18.7 $\mu\text{g/L}$) indicating that the sediments are unpolluted. The As concentration in water samples (average = 8.57-34.7 $\mu\text{g/L}$) considered high. This research first investigated the water qualities and distribution pattern of rare-earth concentration in coastal sediment and water, providing a baseline in the Bay of Bengal, Bangladesh. We have, therefore, drawn new color maps for As, heavy metals and rare-earth using computerized software techniques as *Generic Mapping Tools* (GMT), *Ocean Data View* (ODV) and find the current geochemical pattern. This type of map may be used to establish general baselines against which more specific natural geochemical variations and human-induced perturbations can be appraised.

Keywords: Bay of Bangle, Arsenic (As), Rare-Earth, Sediment, Coastal Water



Stable nitrogen isotopic composition ($\delta^{15}\text{N}$) of dissolved organic nitrogen (DON) in the South China Sea

*Run Zhang¹

1. Xiamen University

Dissolved organic nitrogen (DON) represents an essential N pool, and its stable isotopic composition ($\delta^{15}\text{N}$) may provide unique insights into marine N cycle. However, few results of DON $\delta^{15}\text{N}$ have been reported to date in the global ocean. In this study, we will report DON $\delta^{15}\text{N}$ values in the upper layer waters of the South China Sea (SCS), which is the largest marginal sea west of the Pacific Ocean.

Keywords: South China Sea, $\delta^{15}\text{N}$, DON