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Review Article

Indian Contributions to Chemical Studies in the Indian Sector of Southern Ocean

MELENA A SOARES* and N ANILKUMAR
National Centre for Antarctic and Ocean Research, Earth System Science Organisation, Vasco-da-Gama, Goa 403 804, India

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Southern Ocean is a crucial oceanic regime for understanding the role of biogeochemical cycles in influencing the global climatic changes, as this is a region where signatures of global changes are more pronounced. For an Improved understanding of the link between Southern Ocean processes, biogeochemical cycles and the global climate, scientific research was commenced. Some remarkable research work has been accomplished by Indian Scientific community in the Indian sector of the Southern Ocean (ISSO). A total of six successful multi-disciplinary and multi-institutional expeditions were carried out from the year 2009 to 2015 in the ISSO, with the pilot expedition launched in the year 2004. During the last decade, an understanding of different aspects of the ocean processes has been achieved using chemical studies and proxies. Studies in the ISSO focused on shifts in the oceanic fronts, the carbon and nutrient dynamics, as well as the food web dynamics across the different fronts. The spatial-temporal variation of nutrients, dissolved oxygen, dissolved inorganic carbon and the factors regulating their distribution in the ISSO has also been addressed. Attempts were made to understand the atmosphere-ocean interaction of CO_2 and biogeochemical processes. Some experimental work have been undertaken to assess the nitrogen uptake and influence of micronutrients on phytoplankton. Similarly, stable isotope of oxygen has been used as a proxy in some early studies in the ISSO. Isotopic and molecular investigations have been commenced to understand the biogeochemical processes within the water column, as the biological pump and particulate organic matter (POM) have a major role to play in carbon sequestration and further influencing the global climate variability. Detailed studies on dissolved and particulate organic matter are being carried out to gain knowledge of the biological pump and the factors responsible for climate variability and ocean acidification.

Keywords: Indian Sector of Southern Ocean; Carbon; Oxygen Isotope; Stable Isotopes

Introduction

Southern Ocean (SO) is the largest and one of the most dynamic regions of the world Oceans. This is a regime influenced by the strongest winds and ocean currents. It is characterized by a well-defined frontal structure and supports transport and exchange of heat, salts, and other organic and inorganic components between the major ocean basins, through the Antarctic Circumpolar Current (ACC) (Jasmine *et al*., 2009; Anilkumar *et al*., 2006; Holliday and Read, 1998). The biogeochemical processes in this region have a major role in the global carbon cycle and climate change. The SO is a HNLC (High Nutrients Low Chlorophyll) region with high variability in the hydrographic and chemical characteristics (Martin *et al*., 2013). With a motive to understand this variability and the factors responsible, a pilot expedition in the Indian sector of the Southern Ocean (ISSO) was launched in the year 2004. From 2009 to 2015, six successful multi-disciplinary and multi-institutional expeditions were carried out in the ISSO. Some major national and international scientific institutions/ Universities have participated in these expeditions. With the launch of these expeditions, research in various sectors of oceanographic studies were commenced to understand the biogeochemical processes in the ISSO. Furthermore, the latitudinal variation of calcification depth of foraminifera (*G.bulloides*) was studied using stable oxygen

**Author for Correspondence: E-mail: smelena7@gmail.com*

isotopes. Investigations to identify the sources and processes governing the distribution of Suspended Particulate Matter (SPM) across the different frontal zones in ISSO are also being carried out. Studies addressing the variation of $\delta^{13}C$ and $\delta^{15}N$ of particulate organic matter (POM), and the factors controlling the isotopic signatures of POM in ISSO have been initiated to achieve knowledge of the biogeochemical processes and the biological pump in the ISSO. Some of the processes studied and the findings reported by using chemical proxies in the ISSO and coastal Antarctica have been discussed in this article.

Nutrients and Dissolved Gases

Documenting and understanding the trends of nutrient and dissolved gases, especially, carbon dioxide (CO_2) and CO_2 fluxes is important for providing insights into the biogeochemical processes in the ISSO and their role in the global climate changes. SO is a region characterised by low temperatures, strong winds, surface currents and mixing velocities. The role of temperature and wind stress in the process of CO_2 uptake and variations in CO_2 fluxes is documented in the SO (Anderson *et al*., 2009; Longinelli *et al*., 2012; Waugh, 2014). SO has been demarcated as a major sink of atmospheric CO_2 (Caldeira and Duffy, 2000; Fletcher *et al*., 2006). However, studies have suggested that the strength of SO as a sink is decreasing and the intensity varies seasonally across different zones (Metzl *et al*., 1991; Metzl, 2009). A deeper understanding of the carbonate system and the processes governing CO_2 uptake is of utmost importance to ascertain whether the entire regime acts as a source or sink for atmospheric CO_2. To gain better insights in this aspect, studies on total CO_2 (tCO_2) and partial pressure of CO_2 (pCO_2) in the ISSO were initiated.

A study to address the spatio-temporal variation of pCO_2 and its relation with nutrients and biological production in the ISSO along two transects (48°E and 57°30'E) was taken up during austral summer 2009 (Shetye *et al*., 2012). This study reported low nitrate with higher nutrient utilisation in the Sub tropical front (STF) from 41°-43°S along 48°E and 43°-46°S along 57°30'E. Vertical mixing was assumed to have a major role in the supply of higher tCO_2 to the surface waters. The pCO_2 increased southwards along both the transects and this was attributed to the lower productivity (low TOC and Chlorophyll values) in the Polar Front (PF) region from 48°S to 57°S, during the study period. Based on a correlation analysis of different factors and temperature normalised pCO_2, this study postulated biological processes to be controlling the pCO_2 along 57°30'E transect and physical processes controlling the pCO_2 variation along 48°E transect. They proposed that SO acted as a sink but the area around Crozet Island behaved as a source likely because this region is prone to upwelling. pCO_2 increased southwards mainly due to low biological productivity. This study suggested that southwards the pCO_2 exhibited an increasing trend in the last decade and is associated with the Southern Annular Mode (SAM).

To fill the existing gaps concerning the knowledge of carbon cycle and pCO_2 variability in the ISSO, Shetye *et al.* (2015) studied the physical and biological processes controlling pCO_2 in the surface mixed layer during the transition period from summer to early winter. The authors used a one dimensional model by Louanchi *et al.* (1996), and describe the mixed layer carbon cycle, using in-situ data (January 2009, February 2010, March 2012) and satellite derived data to determine the relative contribution of biological activity, mixing, thermal and air-sea fluxes on pCO_2. Significant spatial and seasonal variation was observed in surface seawater pCO_2 across the frontal zones. An average increase of ~75µatm was reported in this study, from the STF (289 µatm) to the PF (364 µatm). It was also noted that the average pCO_2 estimated in the entire study area increased from 286 µatm in January to 337 µatm in March. A transition from pCO_2saturated waters (March 2009) to under saturated waters (January 2012) was observed during the study. In the month of January, the highest biological drawdown of pCO_2 was reported. This drawdown was associated with biologicalprocesses such as primary production, respiration and calcification, and was considered as the predominant controlling factors as compared to the physical processes like thermal variability, mixing and air-sea fluxes. This study concatenates the previous study (Shetye *et al*., 2012) which addresses ISSO as a sink. The same study also reported that the average nutrients were highest in February, however silicate was maximum in March. A deviation from the Redfield's N/P ratio (16) was displayed with a variation, from an average of 6 to 9 increasing southwards, along the transect. The low N/P ratio indicated higher nitrate utilization. The N/P and Si/N ratios decreased from January to March.

A similar study was carried out in the Antarctic coastal waters and the Enderby basin to address the seasonal biogeochemical dynamics, pCO_2 variability and the air-sea flux through the different seasons (Shetye *et al*., 2016). They found sea-ice plays a crucial role in driving biochemical changes and pCO_2 variability in the coastal Antarctica. Sea-ice changes caused a decrease in sea surface pCO_2 during summer, while it enhances pCO_2 during winter. This study documented a higher pCO_2 in March compared to that in January and February, however, most of the study area was under saturated with pCO_2. Based on the CO_2 fluxes estimated the study inferred, Enderby basin act as a strong sink in January, but a weak sink in other two months and proposed that different factors dominate during different months. The primary productivity, nutrient availability and sea ice were dominating in January but in February temperature played a major role, followed by primary productivity and sea-ice melting. However during March, nutrients, light availability and vertical mixing along with decay of organic matter were significant factors. The study also reported that the regions east of 55°E along the coastal Antarctica acted as a source of CO_2, while the western region acted as a sink for atmospheric CO_2 and associated this deviation with vertical mixing and biological processes.

Studies on nutrient distribution and DO in ISSO waters were initiated by Rajkumar *et al*. (2008). During the pilot expedition to SO, it was reported that the trend in nutrient distribution across the frontal zones matches with the hydrographic properties. The data (nutrient and oxygen) obtained during this study also provided insights of the changes in productivity.

Recently attempts have been made to understand the nutrient variability and nutrient utilisation across the frontals zones in the ISSO using geochemical tracers such as N*(nitrate utilisation) and Si*(Silicate utilisation).

N* and Si* was calculated as follows:

$N^* = NO_3 - 16PO_4$

$Si^* = Si(OH)_4 - NO_3$ (Sarmeinto *et al*., 2004)

It was suggested that the utilization of nitrate was high in the northern region from 39°S to 44°S; whereas the silicate utilisation was higher in the south from 45°S to 52°S (Dessai *et al*., 2011, Tech rep). The study ascertained that the nutrient dynamics across the fronts depends on the physical and biological processes.

Shetye *et al.* (2014) studied silica depletion under the Antarctic sea ice. They hypothesised that diatom bloom driven depletion of dissolved silica in the Antarctic under sea ice waters affect biomineralising organisms. They reported that surface dissolved silica (DSi) increased from subtropics towards the poles and was high all along the coast and also the dominance of diatoms resulted in high sediment organic carbon (3.5 %). They reported intense diatom dominated ice-edge plankton bloom in the Enderby basin causing severe depletion of DSi (< 5 µM) under sea ice. This was supported by the presence of small spicules of sponges under Antarctic ice, also evidence for silica depletion from penecontemperorous dissolution was observed on small style spicules and the authors attributed thelow silica concentration under Antarctic sea ice to diatom bloom which is severely affected by the silicification in sponges.

Stable Isotopic Signatures of Carbon for Understanding CO_2 Exchange

A study to identify the region of CO_2 venting over the SO was carried by Prasanna *et al*. (2015). To achieve this, the atmospheric CO_2 concentration, the $\delta^{13}C$ of atmospheric CO_2, and the $\delta^{13}C$ of DIC was estimated in the sea surface waters across the latitudes in ISSO during 2011, 2012, 2013. They used Keeling's mixing model to trace the source of CO_2. The average $\delta^{13}C$ composition of the source of CO_2 was predicted to be –9.22±0.26 ‰ in the year 2011 and 2012 whereas it was predicted to be 13.49±4.07 ‰ in the year 2013, based on measured data. The end member value for 2011, 2012 was similar but did not match with the expected value of degradation of phytoplankton, thus the authors deduced dissolution of bicarbonates in the water column to be the source of CO_2 during the study period. They also proposed the degeneration of DIC to be favoured in warmer waters. However in 2013, the source of CO_2 was inferred to be degradation of organic matter and mixing of DIC present in seawater. The Keeling's component model identified the end member to have a composition of -36.7 ‰. Another finding from this study was that degassing of CO_2 to the atmosphere was favoured in coastal Antarctic region as this regime experience high wind speed and the surface waters have high DIC.

A recent report by Prasanna *et al.* (2016) addresses the effect of change in water mass properties with depth and latitudinal position on the incorporation of $\delta^{18}O$ and $\delta^{13}C$ in foraminifera (*G.bulloides*). The study also addressed the spatial distribution of $\delta^{13}C$ of DIC in surface waters. The $\delta^{18}O$ and $\delta^{13}C$ values of calcite at equilibrium, was calculated using the temperature and compared it with the isotopic composition of $\delta^{18}O$ and $\delta^{13}C$ in the water column. The measured $\delta^{18}O$ and $\delta^{13}C$ values were higher than the expected values by –2 ‰ and –1 ‰ respectively. They attributed the cause of disequilibrium south of 40°S to a process of mass dependent kinetic fractionation due to deeper depth habitat, partial dissolution, non-equilibrium calcification, oceanic Suess effect and genetic variability. An enrichment in $\delta^{13}C_{(DIC)}$ of surface waters was reported and this was attributed to an increase in organic matter production or its associated removal. Large inter-annual variation between the period of 2012 and 2013 was documented. They also proposed a model for production and export of organic matter. This model can be used to simulate $\delta^{13}C_{(DIC)}$ values for biologically productive regions in the SO. The model accounts for the $\delta^{13}C_{(DIC)}$ values in a given location based on the production rate of organic carbon, total organic carbon and the removal rate of organic carbon, it also demonstrates that a steady state of the carbon isotope ratio of water can be achieved in a relatively short time of ~5000 days. Beyond 50°S, the $\delta^{13}C_{(DIC)}$ values were low. The study ascertained that these $\delta^{13}C_{(DIC)}$ values are controlled by the nutrient supplied by Antarctic Bottom Water and upwelling.

Use of Stable Isotope of Oxygen ($\delta^{18}O$) and Hydrogen (δD) as a Proxy

$\delta^{18}O$ is generally used as a tracer to identify different water masses. δD values of seawater are sensitive to various processes of the hydrological cycle like evaporation, precipitation, and freezing. Therefore, study of δD and $\delta^{18}O$ along with salinity are unique and important to understand ocean surface processes.

A study was carried out by Tiwari *et al.* (2013) by coupling stable isotopes of oxygen and salinity in surface sea water to ascertain the ocean-atmospheric processes across the different fronts in the ISSO as well as to identify the water masses in this region. The study highlighted the relation between $\delta^{18}O$ and sea surface salinity pertaining to distinct water mass and confirmed the prevalence of evaporation/precipitation. The subtropical zone was dominated by evaporation/precipitation, whereas in the Antarctic zone, ice melting-freezing is dominant. The effect of continental ice melt was noted at the southernmost location (66.73°S). Using this stable isotope proxy the Subtropical and Polar fronts were identified at 44°S and 54°S respectively. This was succeeded by a study (Tiwari *et al.*, 2015) to document the $\delta^{18}O$ variability, the relation of $\delta^{18}O$ with salinity across the fronts in ISSO. The study also identified the origin, of the waters in the eddy encountered area. The slope of $\delta^{18}O$-salinity relation indicated that the warm core eddy was formed in a region dominated by evaporation and precipitation, while the waters surrounding the eddy are from a region dominated by freezing and melting. The role of eddies in the formation of intermediate waters in the SO was also explored in this study. The Antarctic Intermediate Waters (AAIW) with $\delta^{18}O$ values of 0.0 ‰ to 0.2 ‰, flowing below the Antarctic Surface waters which displayed the lowest temperature, salinity and $\delta^{18}O$ values (–0.4 ‰ to –0.2 ‰) were identified. The AAIW forms in the polar frontal waters, a region with the dominance of freezing/melting, however it was observed at a shallower depth which was possibly due to the influence of the eddy.

Experimental Work

To understand the nitrogen uptake and its role in limiting phytoplankton productivity, a few experiments have been carried out in the ISSO. Earlier studies have ascertained iron limitation in the SO and an addition of iron can increase primary productivity (Geider & Rocha, 1994; Hutchins *et al.*, 1995). However the effect on different N-substrate was unknown. One such initiative was a bottle scale ^{15}N tracer iron experiment carried out by Prakash *et al.* (2010) to assess addition of iron on the N-uptake rate and f-ratio in the ISSO. It was reported that iron addition not only enhance NO_3 uptake but also showed an increase in ammonia and urea uptake with insignificant effect on the f-ratio. The addition of iron did not enhance N-uptake based primary production at the initial stage but increased the uptake at a later stage.

Another mesocosm experiment was carried out by Dessai *et al*. (2011, Tech. Rep) to study the influence of iron and cobalt on phytoplankton community structure in the subtropical and polar front of the ISSO. It was reported that individually both the micronutrients stimulated growth when added individually but when added in combination it did not trigger much growth. It was also observed that diatom sustained longer in a cobalt enriched environment whereas the biomass was higher in the iron enriched environment.

Suspended Particulate Matter (SPM)

A study was taken up by Dessai *et al*. (2011) to gain insights into the spatial distribution of SPM and the associated trace metals and to identify its sources. The authors reported high SPM and low salinity in surface waters and also high SPM at 1000 m along with high iron and manganese in Antarctic zone. The study suggested that the particulate matter in the northern region especially the subtropical front was mainly of biogenic origin, with enrichment of calcium. It also stated that, temperature and salinity controlled primary production, has a major role in the SPM distribution of this region. Conversely, in the southern region SPM was of terrigenous origin and rich in iron and manganese. The inputs to the SPM in Southern region was proposed to be mainly from melt water and from a region with low productivity dominated by diatom. A similar study was carried out to understand the processes controlling the distribution of SPM across the frontal zones during austral summer 2010, wherein it was reported that the highest SPM was quantified in the Subantarctic zone followed by the Antarctic zone and this variability was attributed to the different biogeochemical processes dominating in the different zones of ISSO (Nayak & Noronha, Tech. rep., 2011)

Particulate Organic Matter (POM) in the Water Column

The ISSO being characterised by different oceanic fronts and influenced by eddies, this region is highly dynamic in terms of air-sea flux of CO_2 as well as the factors governing the fate of organic matter (production, remineralisation) and preservation. POM is a key component of the biological pump and it is known from earlier studies, that a major portion of POM is remineralised in the upper photic layer (Honjo *et al*., 2008; Duforet-Gauier *et al*., 2010). Therefore to understand the biogeochemical processes isotopic and molecular studies are important. The stable isotopic ratio of carbon and nitrogen of POM ($\delta^{13}C$ and $\delta^{15}N$) are often used to understand the conditions prevailing at the time of carbon fixation and to get insights of the processes related to organic matter cycling (Lourey *et al*., 2003, 2004 & references within). Recently investigations to understand the biogeochemical cycle of POM using isotopic and molecular studies have been initiated.

One such study addressed the variation of $\delta^{13}C$ and $\delta^{15}N$ of POM, and the factors controlling the isotopic signatures of POM in surface waters in the ISSO (Soares *et al*., 2015). In this study it was reported that in the ISSO, physical processes like temperature gradient and eddies as well as the biological processes contribute equally in the isotopic characterisation of POM, and these factors are responsible for the variation in $\delta^{13}C$ and $\delta^{15}N$ across the fronts in ISSO surface waters. During austral summer 2012, the $\delta^{13}C$ of POM in Subtropical front was enriched in $\delta^{13}C$ (average –22.81‰), relative to the $\delta^{13}C$ of colder Polar Front waters that displayed depleted values (average –25.82‰) similar to $\delta^{15}N$ that was enriched in the Subtropical front. It was postulated that the community structure had a major role in determining the isotopic signatures of POM with the dominance of dinoflagellates in the eddy prone, low nutrient Subtropical front waters with $\delta^{13}C$ enriched POM and diatoms dominated nutrient rich colder water displaying depleted $\delta^{13}C$ of POM.

Other Related Studies

Use of $\delta^{18}O$ as a Proxy in Planktonic Foraminifera Studies

Saraswat and Khare (2010) tried to ascertain the water depth at which the planktonic foraminiferal spp. *G. bulloides* calcifies its shell based on the stable isotopic composition of oxygen ($\delta^{18}O$) in its shells. A comparison of seawater salinity and temperature estimated from $\delta^{18}O$ of *G. bulloides* $\delta^{18}O$ show that calcification depth of *G. bulloides* varies latitudinally.

The study also stated that north of 43°S the calcification depth was deeper approximately 200 m, determined from the estimated temperature well matching the seawater temperature at 200 m. However, south of 43°S the calcification depth was shallower.

Subsequently, a study was carried out to evaluate the spatial variability of isotopic value of foraminifera across different frontal regimes (Tiwari *et al*., 2011). This study documented, planktonic foraminifera secrete their shell at isotopic equilibrium with seawater. Also, comparative analysis of $\delta^{18}O_{calcite}$ and predicted values $\delta^{18}O_{calcite}$ revealed a similar trend as that measured for the shells. They reported $\delta^{13}C$ values of plankton net samples and core top sediment samples increased southwards and attributed it to the productivity fluctuation which increase southwards due to the influx of nutrients from ice melt, since $\delta^{13}C$ variability appear to be governed mostly by primary productivity.

Recently, Mohan *et al*. (2015) demonstrated that there is a shift in the abundance of foraminifera from Subtropical Front to Polar Front and reported $\delta^{18}O$ in sediments is higher compared to towed plankton samples. The study proposed that some part of the shell precipitates in cooler deeper waters where secondary calcification occurs. This study suggested the absence of dissolution in the foraminifera from surface sediments and attributed it to low organic biomass.

Future Prospects

Although many studies have been carried out in the ISSO, the region is still understudied in terms of the biogeochemical processes. It is of utmost importance to understand the biogeochemical processes in the region as the SO have a strong global influence. Most important being the CO_2 dynamics, although studies are going on, they need to be extended on a larger scale and the system needs to be monitored to quantify the fluxes. It is essential to address Ocean acidification, which significantly influences the ocean biological community structure and other biochemical processes, consequently affecting the food web dynamics. Besides, detailed studies to understand the biological pump and processes like remineralisation within the water column are crucial, as the water masses formed in the ISSO can upwell at a different location causing degassing/flux of CO_2 to the atmosphere. As the carbon, nutrients and food web dynamics are all inter-linked and involve a complex interplay of varied physical, biological and biogeochemical processes which need to be studied in detail to understand and further predict the global climate variability.

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