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SEASONAL VARIATIONS OF TOTAL ^{234}Th AND DISSOLVED ^{238}U CONCENTRATION ACTIVITIES IN SURFACE WATER OF BRANSFIELD STRAIT, ANTARTICA, FROM MARCH TO OCTOBER 2011

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ABSTRACT

In this study the naturally occurring radionuclides ^{234}Th and ^{238}U were used to investigate the magnitude of upper ocean particulate organic carbon export in Bransfield Strait, Southern Ocean. This region is the largest oceanic high-nitrate low-chlorophyll (HNLC) area in the world and is known to contribute to regulate of the atmospheric CO_2 via the biological pump. Due to its different geochemical behavior in seawater, the resulting U/Th disequilibria can be easily used to constrain the transport rates of particles and reaction processes between solution and particulate phases. Sampling occurred during the summer (March and November) 2011. Total ^{234}Th activities in surface seawater samples ranged from 1.3 to 3.7 dpm L^{-1} (station EB 011) during March/11 campaign, while in October/11 total ^{234}Th activity concentrations varied from 1.4 to 2.9 dpm L^{-1}. Highest total ^{234}Th activities were found late in the austral summer season. Activity concentrations of dissolved ^{238}U in surface seawater varied from 2.1 to 2.4 dpm L^{-1}. Taking into account all sampling stations established in March and October/11 the relative variability of total ^{234}Th distribution was 22%.

1. INTRODUCTION

To study carbon cycle, scientists apply ^{234}Th, a natural radionuclide of ^{238}U series, as a tracer of the flux of particulate organic carbon (POC) exported in the ocean [1]. The natural radionuclides have been used as tracers in the investigation of ocean processes and managing the coastal region. Their applications in oceanographic works and/ or marine pollution have helped scientists to elucidate the processes that occur in the water column (e.g. particle transport, carbon cycle, biogeochemical cycles, scavenging) or in sediments (sedimentation, deposition accumulation, transport and upwelling).

The Southern Ocean plays an important role in global climate change being considered a sink of atmospheric carbon dioxide (CO_2). It is the largest repository of available macronutrients in surface waters and a very important region for the formation of intermediate and deep waters [2]. According to several authors, the Bransfield Strait is a highly productive shelf area for all trophic levels, and considered a very dynamic region under the influence of Weddell and Belingshausen Seas [3-6]. There are currently few direct measurements of particulate export in the Bransfield Strait, Antartica. Such knowledge has proven to be essential for

predicting the fate of organically bound or biologically active contaminants, such as anthropogenically produced CO_2, hydrocarbons and metals [7]. The predominant transfer of these contaminants from the upper ocean to underlying sediments is generally thought to be biologically mediated.

The primary mechanism for removing CO_2 from the atmosphere into the ocean is "biological pump", governed by photosynthesis process by which about 20 – 40 % of fixed carbon by phytoplankton is exported to the ocean bottom for the sinking biogenic material [8, 9]. The seaweeds present in the Southern Ocean is the most important group of primary producers, converting inorganic carbon into organic matter through photosynthesis, making it the basis of the food chain. Therefore, phytoplankton plays a key role in biogeochemical cycles and in the transfer of matter and energy to the environment.

^{234}Th is a naturally occurring particle-reactive radionuclide which has been commonly used to study particle scavenging in the upper ocean [10]. Since the half-life of ^{234}Th is 24.1 days, the disequilibrium between its soluble conservative parent ^{238}U and the measured ^{234}Th activity reflects the net rate of particle export from the upper ocean on time scales of days to weeks. This period of time makes ^{234}Th a useful radionuclide to study POC fluxes exported in the Southern Ocean during Austral Summer phytoplankton bloom. This methodology is based on the assumption that a decrease of $^{234}Th/^{238}U$ activity ratio implies a increasing of the flux of particulate matter [10]. In the surface ocean, both the formation of fresh particle surfaces (proportional to primary production) and the packaging of particles into sinking aggregates (export) are reflected in the observed ^{234}Th distribution in the marine environment.

The uranium cycle in the ocean is in steady state, with a residence time of 400.000 years, substantially greater than the average time of oceanic mixture [11]. Under oxidizing conditions, uranium in seawater occurs predominantly as a stable ion of uranyl carbonate $UO_2(CO_3)_3^{-4}$. As ^{238}U is conservative in seawater, its activity varies very little as a function of depth and is proportional to salinity.

This study evaluated the seasonal variations of both ^{234}Th and ^{238}U distributions in surface waters of the Bransfield Strait, during March (OPERANTAR XXIX) and October (OPERANTAR XXX) campaigns.

2. MATERIAL AND METHODS

2.1. Study Area

The study area is located in the Bransfield Strait, which is considered a semi-enclosed sea approximately 300 km long and 100 km wide, situated between the South Shetland archipelago and northeastern tip of the Antartic Peninsula. The region is considered a narrow, volcanic and seismically active extensional basin. The northeastern part of the Bransfield Strait opens in the Scotia and Weddell Seas. And the southwest part opens in the Bellingshausen Sea through the Gerlache Strait [5, 6, 12].

The bottom topography of the Bransfield Strait consists of a central basin deeper than 1000 m. The shelf slope waters around the South Shetland Islands consist of the water strong influenced by local processes of heating and freshwater runoff referred to as the Shelf Slope Water, and the intrusion water from the Antartic Circumpolar Current deep water which intrudes into the Bransfield Strait from the deep through between Brabant and Smith Islands and remains near the South Shetland Islands. At the shelf slope, a western boundary current, the Bransfield Current, determines the water export from the Bransfield Strait through the shelf slope of the South Shetland Islands and transport nutrients, metals and biota [5, 6].

2.2. Sampling

Sampling cruises were carried out onboard R/V *Ary Rongel*, covering an area located between latitudes 63º33'25"S - 60º27'99"S and longitudes 53º00'49"W - 62º09'47"W. The first sampling was performed late in the 2010 Austral Summer, between 8th March to 6th April 2011, in which a total of 47 surface stations were occupied (Fig.1).

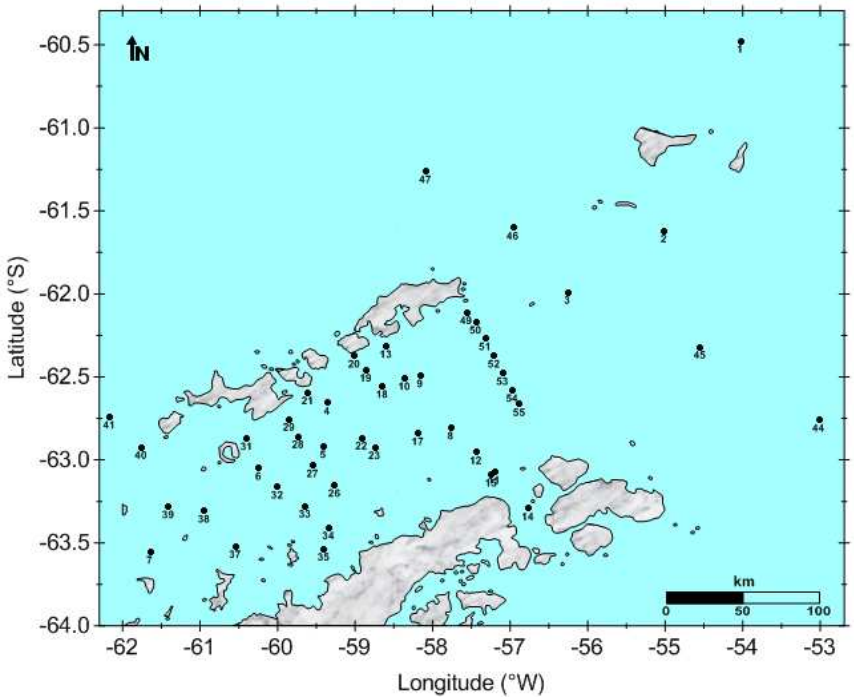

Figure 1: Location map of the surface water stations sampled during March-April/2011 campaign (OPERANTAR XXIX) in the Bransfield Strait, Antartica.

The second sampling campaign was carried out early in 2011 Austral Summer, between 13th October to 14th November 2011, in which a total of 17 surface stations were occupied (Fig.2).

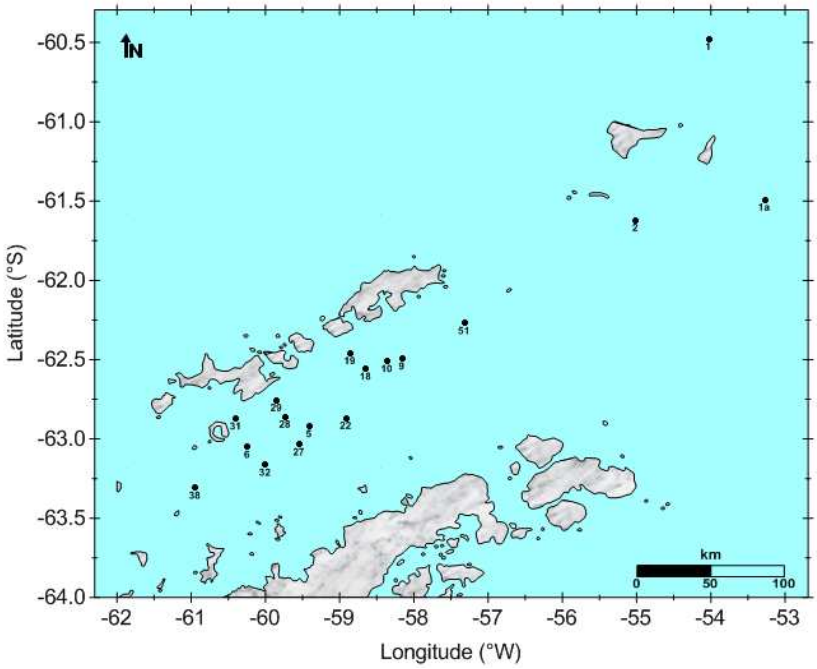

Figure 2: Location map of the surface water stations sampled during October-November/2011 campaign (OPERANTAR XXX) in the Bransfield Strait, Antartica.

Seawater samples were collected using Rosette and CTD coupled, obtaining the record of temperature and salinity in continuous mode. The depth records were obtained with an XBT profile meter.

In each hydrochemical station nutrients were also sampled to examine pathway sources. For such purposes, seawater samples were transferred in sample-rinsed polyethylene bottles, and filtered through HCl-washed Wathman GF/F filters immediately upon sample collection. Those samples were kept frozen until return of the *RV Ary Rongel* to Brazil, where they were assayed in the chemical laboratory of the Oceanographic Institute of the University of São Paulo (Labnut) for nitrate, ammonia, silica and inorganic phosphate analyses following recommended procedures [13-15].

2.3. Total ^{234}Th analysis

The measurement of total (dissolved + particulate) ^{234}Th activity in seawater samples was based on the MnO_2 precipitation method described by Rutgers Van der Loeff & Moore [1]. The technique involves formation of a precipitate of manganese dioxide (MnO_2), which drags preferably by co-precipitation ^{234}Th and insignificant amount of ^{238}U. Measurements of ^{234}Th were performed by detection of its decay product ^{234m}Pa which is a beta emitter of high energy [16, 17]. Analyses of total ^{234}Th activities were performed at the Environmental Radiometrics Laboratory of IPEN/CNEN-SP using a gas-flow low background proportional counter from EG&G Berthold, model LB 770.

For this analysis, a two-liter sample of seawater was collected without filtering in which reagents such as potassium permanganate and manganese chloride ($MnCl_2$ and $KMnO_4$) were added to form MnO_2 precipitate. ^{229}Th standard solution was added as a yield monitor. The

precipitate was set aside for 8 - 16 hours and then filtered through filter paper of fiber glass with 47mm diameter (1.2μm pore size). This methodology might be implemented easily in marine samples with accuracy and precision ≤ 5% [16, 17].

Initial count rates were typically 2-4 counts per minute (cpm) for total ^{234}Th. The counting was repeated over a 6-month period (> six ^{234}Th half-lives). This was to check that the activity decrease followed the decay of ^{234}Th and also allowed a background correction for activity intrinsic to the detector and from other long-lived natural beta-emitters. The final counting rates averaged 0.5 cpm relative to the detector backgrounds of 0.4-0.6 cpm [17].

The methodology also involved the determination of ^{229}Th to estimate the chemical yield of the process. Anion exchange chemistry was performed and recovery of added ^{229}Th yield monitor was quantified by inductively coupled plasma-mass spectroscopy. Corrections were applied to ^{234}Th activity calculations on the basis of ^{229}Th recoveries. All data are decay corrected to the time of collection and reported with a propagated error that includes uncertainties associated with counting, sample volumes and other calibration errors. Errors were typically ± 0.04-0.06 cpm L^{-1} on total ^{234}Th [17].

2.4. Dissolved ^{238}U estimation

The dissolved ^{238}U activity must be known to estimate the ^{234}Th flux exported [10]. As ^{238}U is conservative in seawater, its activity varies very little as a function of depth and it is proportional to salinity according to the following equation [11]:

$$^{238}\text{U (dpm L}^{-1}) = 0{,}069 \text{ x salinity} \quad (1)$$

3. RESULTS AND DISCUSSION

Temperature distribution maps of surface seawater collected at OPERANTAR XXIX and OPERANTAR XXX are presented in Figures 3 and 4, respectively. Temperature data varied from -0.2ºC to 2.6ºC (March/2011) and from -2.1 ºC to 0.6 ºC (October/2011). Colder seawater samples were detected around Trinity Peninsula and Nelson Island during the end and beginning of Austral Summer, respectively. Relatively warm waters were observed South Shetland Islands at the final summer season, while at the summer beginning higher water temperatures were found closest to Elephant Island and central Bransfield Strait, around Livingston Island.

Salinity distribution maps of surface seawater collected at OPERANTAR XXIX and OPERANTAR XXX are presented in Figures 5 and 6, respectively. Results showed a variation from 33.70 to 34.54 in March/2011, while in October/2011 salinity values ranged from 33.45 to 34.16. The lowest salinity values were reported at the South Shetland Islands during the beginning and end of Austral Summer, respectively. Highest salinities were measured around Trinity Peninsula at the final summer season, and at the next summer beginning closer to Elephant Island and central portion of Bransfield Strait.

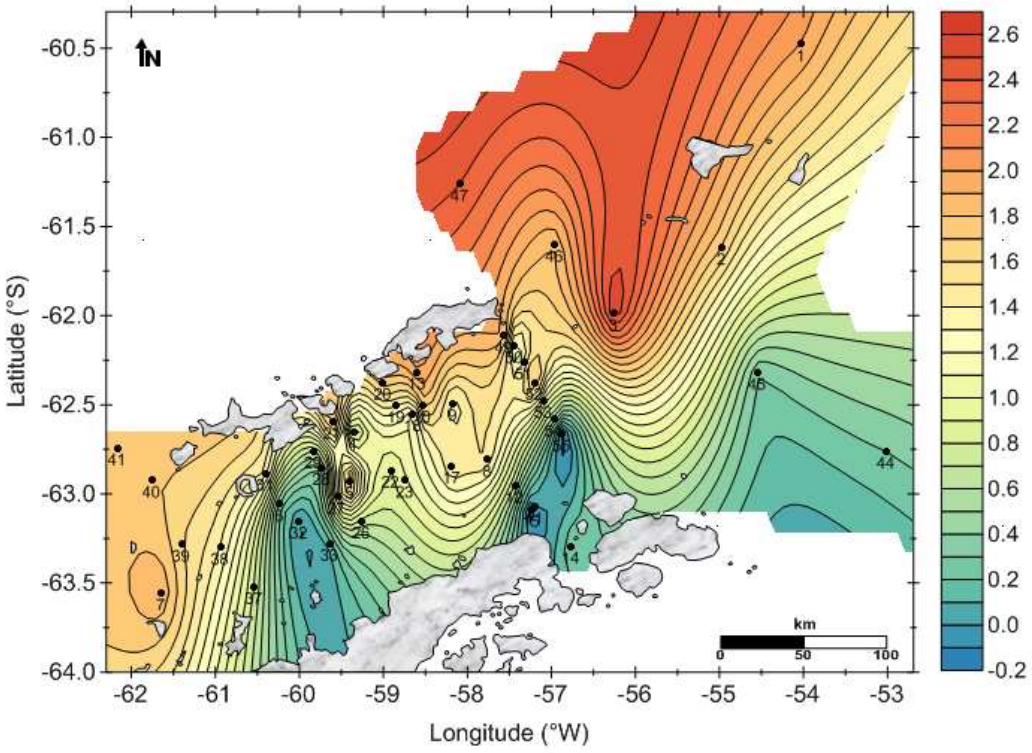

Figure 3: Temperature distribution map of surface seawater collected at OPERANTAR XXIX expedition.

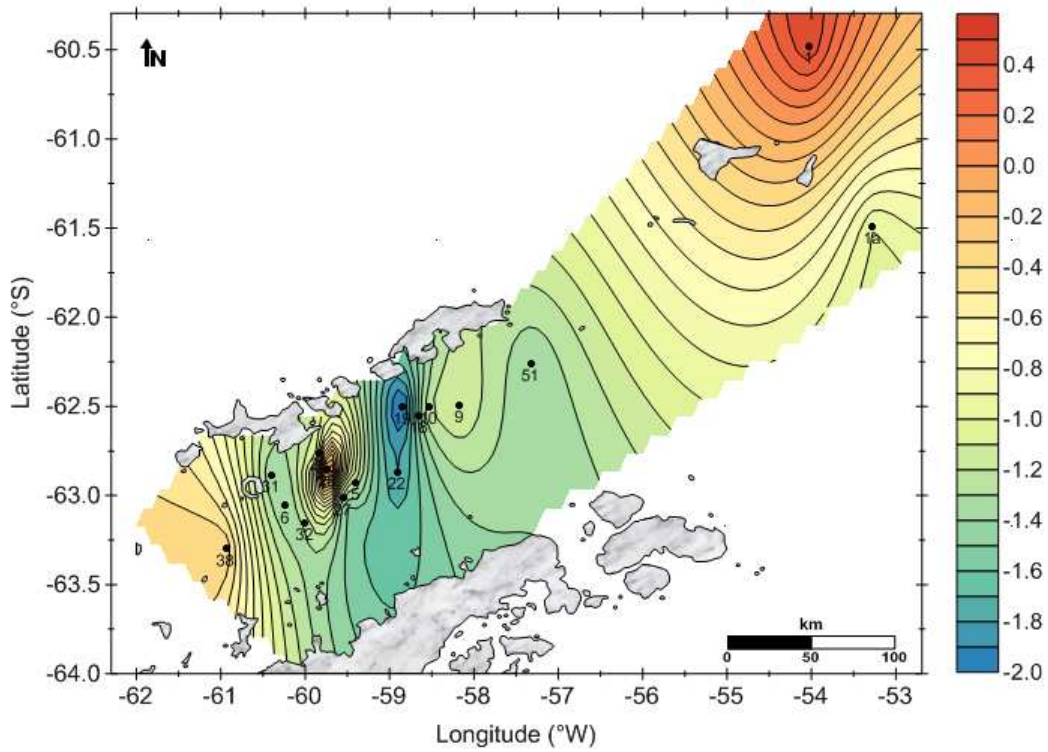

Figure 4: Temperature distribution map of surface seawater collected at OPERANTAR XXX expedition.

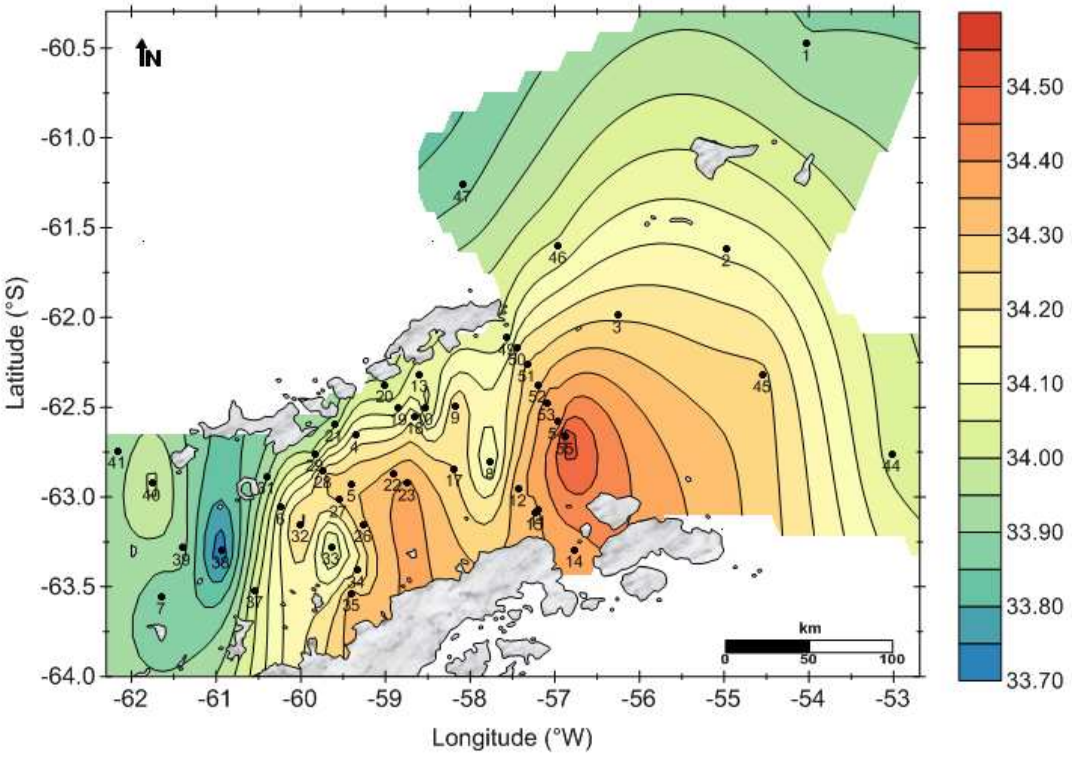

Figure 5: Salinity distribution map of surface seawater collected at OPERANTAR XXIX expedition.

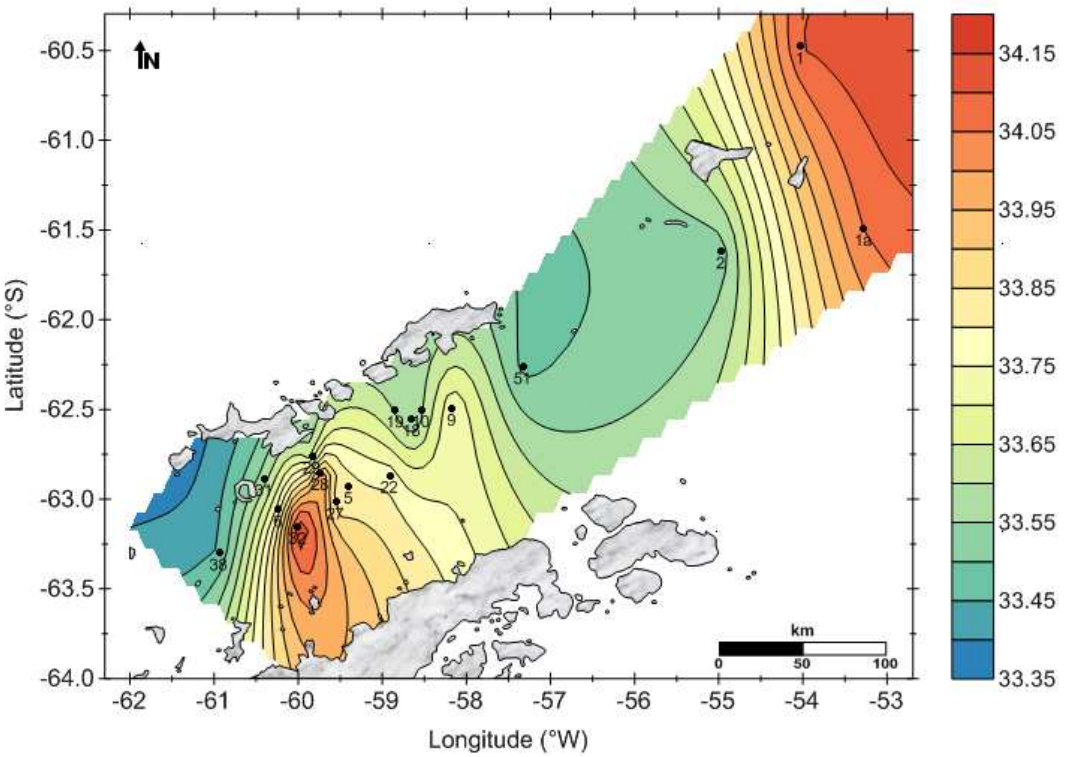

Figure 6: Salinity distribution map of surface seawater collected at OPERANTAR XXX expedition.

According to Tokarczyk [3] and Grácia *et al*. [5], waters relatively colder with higher salinity values observed around Trinity Peninsula during the end of summer season indicate intrusion of waters from Weddell Sea, while warmer less saline waters detected in South Shetland Islands show influence of Bellinghausen Sea.

Activity concentrations of total ^{234}Th, dissolved ^{238}U, total ^{234}Th/^{238}U activity ratios and salinity of surface seawater samples from OPERANTAR XXIX and OPERANTAR XXX expeditions are presented in Tab. 1 and 2, respectively.

Total ^{234}Th activities in surface seawater samples studied along the Bransfield Strait ranged from 1.3 dpm L^{-1} to 3.7 dpm L^{-1} (station EB011) during March/2011 campaign, while in October/2011 total ^{234}Th activity concentrations ranged from 1.4 dpm L^{-1} to 2.9 dpm L^{-1} (station EB006) (Figure 7 and 8).

Activity concentrations of dissolved ^{238}U in surface seawater were estimated from salinity data, using equation 1. Dissolved ^{238}U activities determined in surface seawater sampled in March/2011 and October/2011 varied from 2.3 dpm L^{-1} to 2.4 dpm L^{-1}, the highest values being observed at station EB011 and EB032, respectively (Figure 9 and 10).

Activity ratios of total ^{234}Th/^{238}U up to 1.52 (station EB011) were observed during March/2011 campaign, while in October/2011 this activity ratio varied between 0.60 and 1.23 (the higher value at station EB006) (Figure 11 and 12).

Non-conservative increases of total ^{234}Th activities related to dissolved ^{238}U (resulting in AR > 1.0) were observed in 14 surface hydrochemical stations performed in March/2011, while in October/2011 this happened in 7 locations. These increases can be representative of several processes, among them: (a) occasional lower resolution of total ^{234}Th measurements; (b) processes of re-suspension and /or upwelling; (c) additional sources of ^{234}Th to the studied scenario, as for example, those related to the ice melting and runoff.

Once ^{234}Th is a particle-reactive radionuclide useful to study particle scavenging in the upper ocean and this methodology is based on the assumption that a decrease of ^{234}Th/^{238}U activity ratio implies a increasing of the flux of particulate matter, activity ratios of total ^{234}Th/ ^{238}U close to 1.0 indicate lower fluxes of particulate matter exported from surface waters. Values around 0.6 like those observed in the euphotic zone at stations EB001, EB004, EB006, EB007, EB010, EB021, EB029, EB037, EB038, EB039, EB040 and EB041 in March/2011 suggest higher fluxes of particulate matter exported. During October/2011 expedition this was also verified at stations EB027, EB028 and EB029, respectively.

Table 1: Activity concentrations of total ^{234}Th, dissolved ^{238}U, total ^{234}Th/^{238}U activity ratios and salinity of surface seawater samples from OPERANTAR XXIX expedition.

Station	Salinity	Total ^{234}Th (dpm L^{-1})	^{238}U (dpm L^{-1})	^{234}Th/^{238}U
EB001	33.91	1.7	2.3	0.71
EB002	34.14	2.2	2.4	0.93
EB003	34.24	2.5	2.4	1.07
EB004	34.17	1.8	2.4	0.75
EB005	34.30	2.0	2.4	0.84
EB006	34.13	1.6	2.4	0.68
EB007	33.86	1.8	2.3	0.76
EB008	34.09	2.5	2.4	1.06
EB009	34.25	2.7	2.4	1.15
EB010	34.20	1.8	2.4	0.74
EB011	35.35	3.7	2.5	1.52
EB012	34.37	2.2	2.5	0.94
EB013	34.08	2.3	2.4	0.99
EB014	34.36	2.3	2.4	0.95
EB015	34.32	2.5	2.4	1.04
EB017	34.25	2.4	2.4	1.02
EB018	34.22	2.7	2.4	1.16
EB019	34.07	2.5	2.4	1.05
EB020	34.01	2.1	2.4	0.89
EB021	33.96	1.5	2.3	0.65
EB022	34.32	2.1	2.4	0.87
EB023	34.37	2.0	2.4	0.86
EB026	34.30	2.1	2.4	0.90
EB027	34.27	2.2	2.4	0.91
EB028	34.27	2.1	2.4	0.86
EB029	33.99	1.8	2.4	0.78
EB031	33.94	2.0	2.3	0.86
EB032	34.29	2.1	2.4	0.88
EB033	34.00	2.1	2.4	0.87
EB034	34.16	2.2	2.4	0.95
EB035	34.34	2.1	2.4	0.90
EB037	34.05	1.3	2.4	0.57
EB038	33.70	1.4	2.3	0.59
EB039	33.93	1.7	2.3	0.72
EB040	34.02	1.8	2.4	0.76
EB041	33.92	1.4	2.3	0.60
EB044	34.03	2.5	2.4	1.07
EB045	34.26	2.5	2.4	1.04
EB046	34.06	2.0	2.4	0.85
EB047	33.89	2.1	2.3	0.89
EB049	34.07	2.0	2.4	0.85
EB050	34.21	2.6	2.4	1.10
EB051	34.26	2.6	2.4	1.08
EB052	34.26	2.3	2.4	0.98
EB053	34.41	2.4	2.4	1.01
EB054	34.33	2.2	2.4	0.92
EB055	34.54	2.8	2.4	1.16

Table 2: Activity concentrations of total ^{234}Th, dissolved ^{238}U, total ^{234}Th/^{238}U activity ratios and salinity of surface seawater samples from OPERANTAR XXX expedition.

Station	Salinity	Total ^{234}Th (dpm L^{-1})	^{238}U (dpm L^{-1})	^{234}Th/^{238}U
EB001	34.10	2.3	2.4	0.98
EB001α	34.05	2.4	2.4	1.00
EB002	30.53	1.9	2.1	0.91
EB005	33.82	2.2	2.3	0.94
EB006	33.77	2.9	2.3	1.23
EB009	33.74	2.2	2.3	0.93
EB010	33.59	2.2	2.3	0.96
EB018	33.55	2.1	2.3	0.90
EB019	33.59	2.3	2.3	1.00
EB022	33.78	2.5	2.3	1.05
EB027	33.81	1.8	2.3	0.79
EB028	34.01	1.8	2.4	0.76
EB029	33.62	1.4	2.3	0.60
EB031	33.58	2.5	2.3	1.06
EB032	34.16	2.4	2.4	1.02
EB038	33.46	2.4	2.3	1.05
EB051	33.49	2.3	2.3	0.98

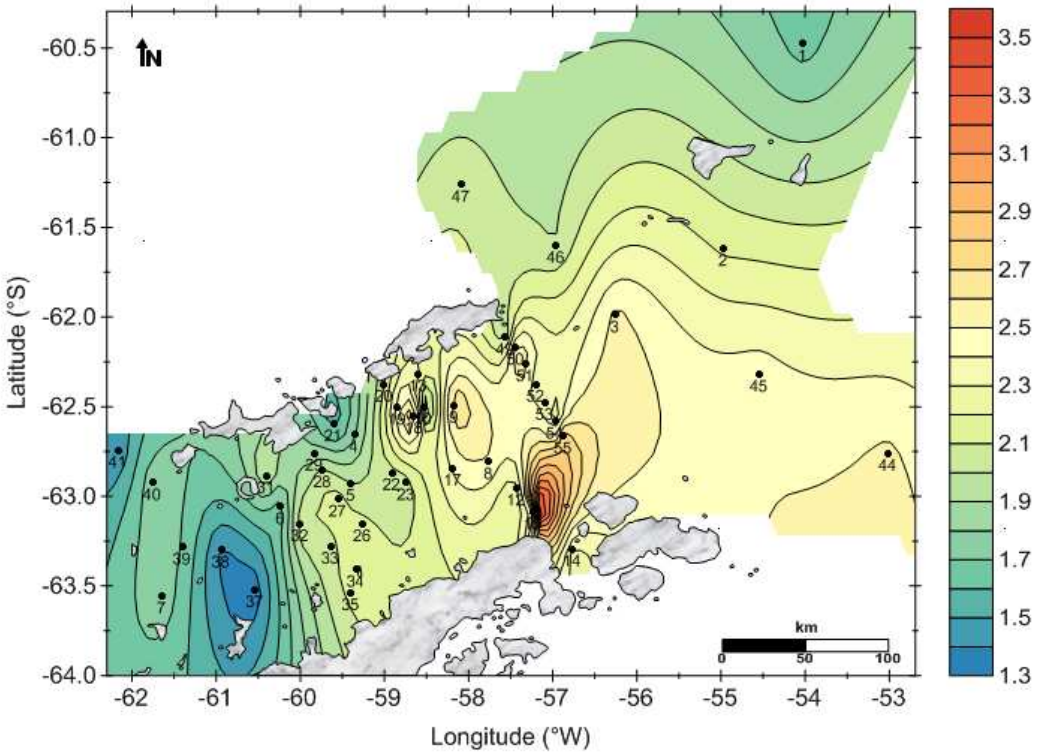

Figure 7: Total ^{234}Th (dpm L^{-1}) distribution map of surface seawater collected at OPERANTAR XXIX expedition.

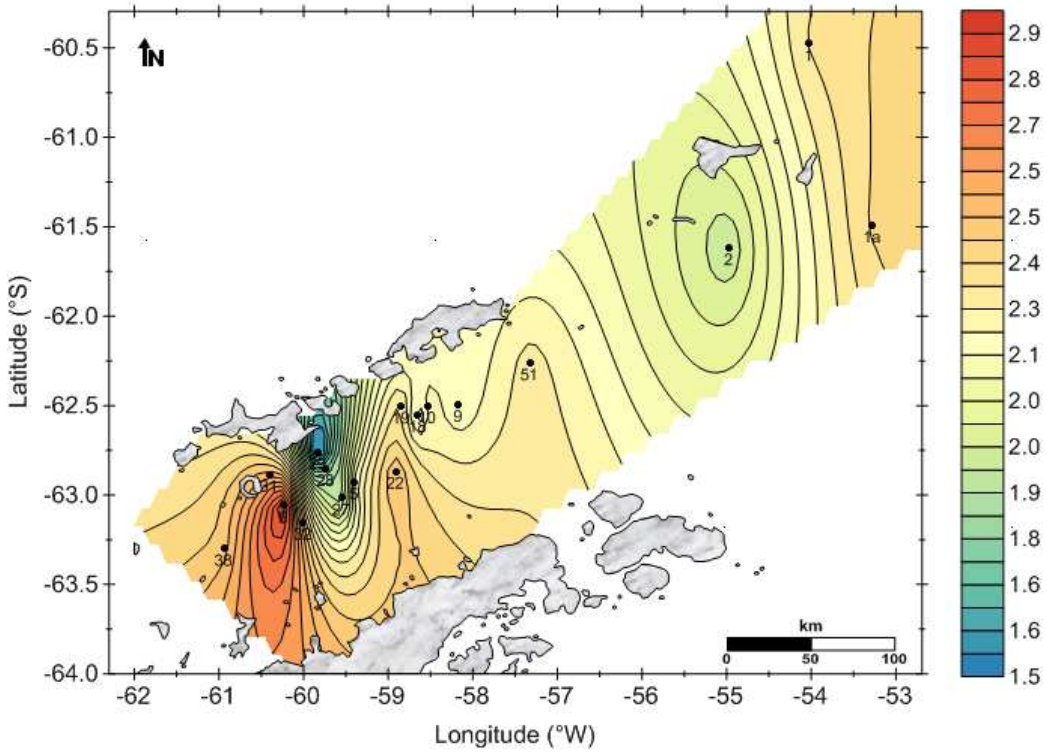

Figure 8: Total ^{234}Th (dpm L^{-1}) distribution map of surface seawater collected at OPERANTAR XXX expedition.

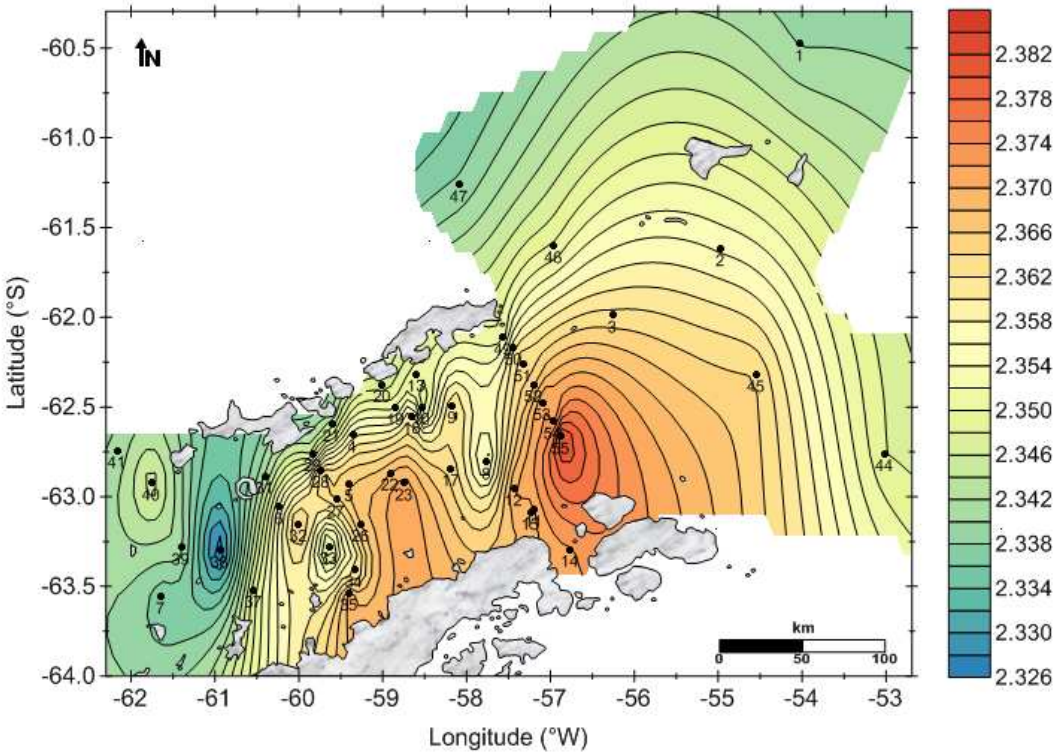

Figure 9: Dissolved ^{238}U (dpm L^{-1}) distribution map of surface seawater collected at OPERANTAR XXIX expedition.

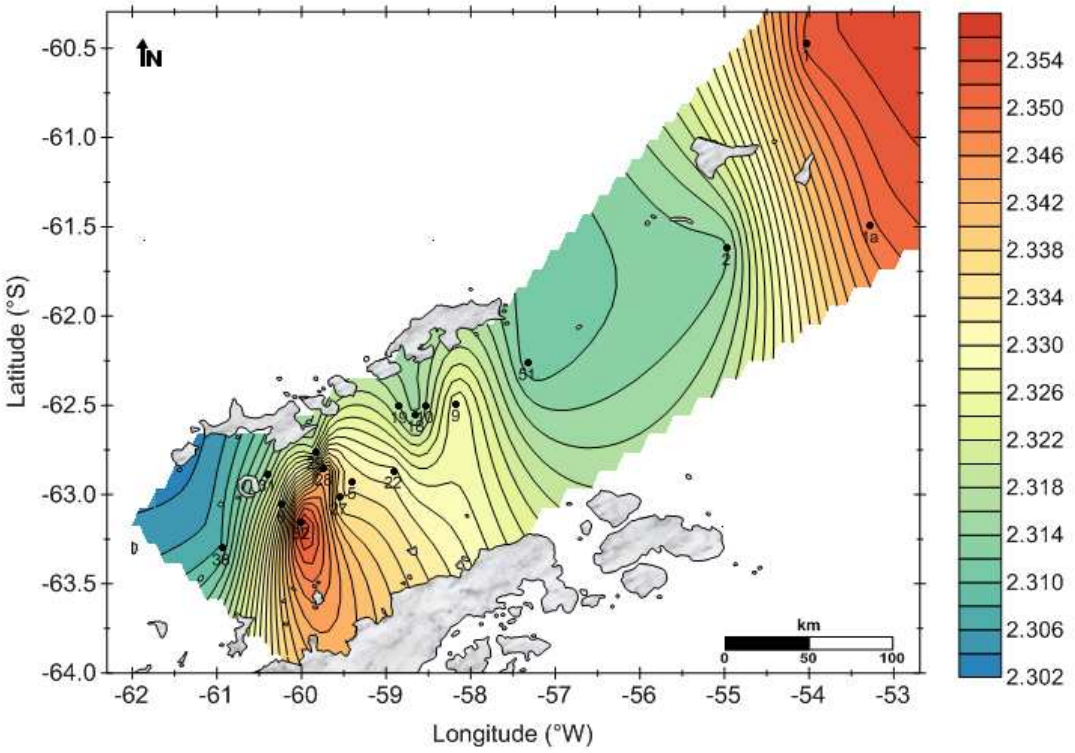

Figure 10: Dissolved ^{238}U (dpm L^{-1}) distribution map of surface seawater collected at OPERANTAR XXX expedition.

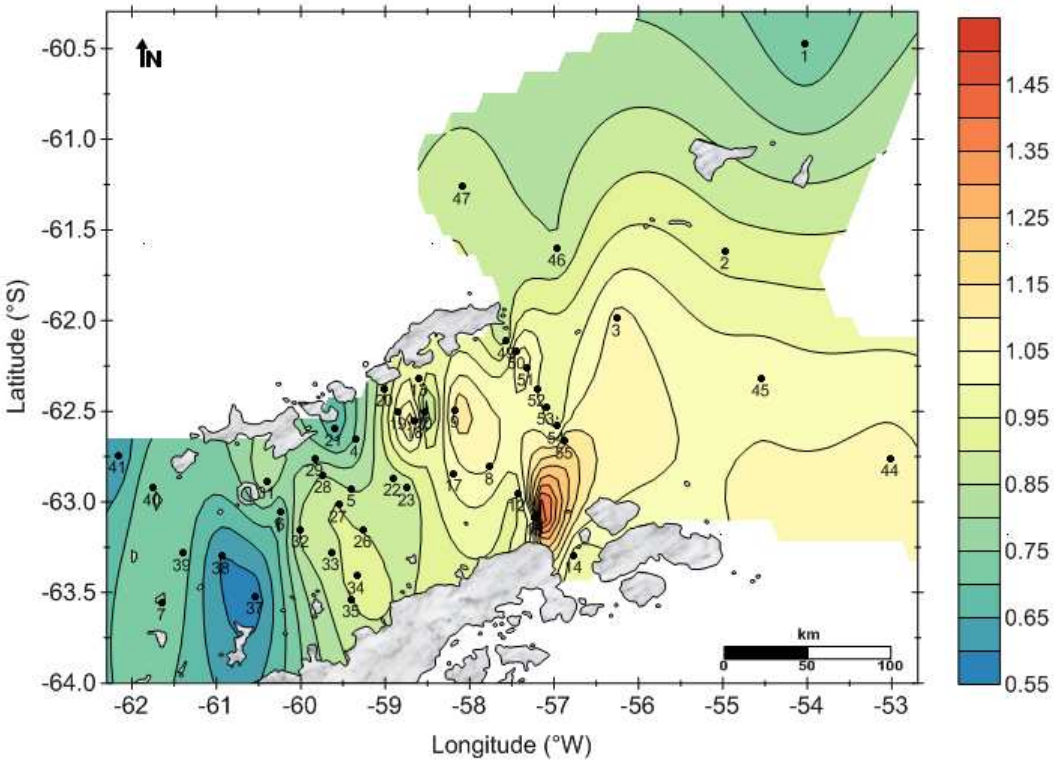

Figure 11: Total $^{234}Th/^{238}U$ AR distribution map of surface seawater collected at OPERANTAR XXIX expedition.

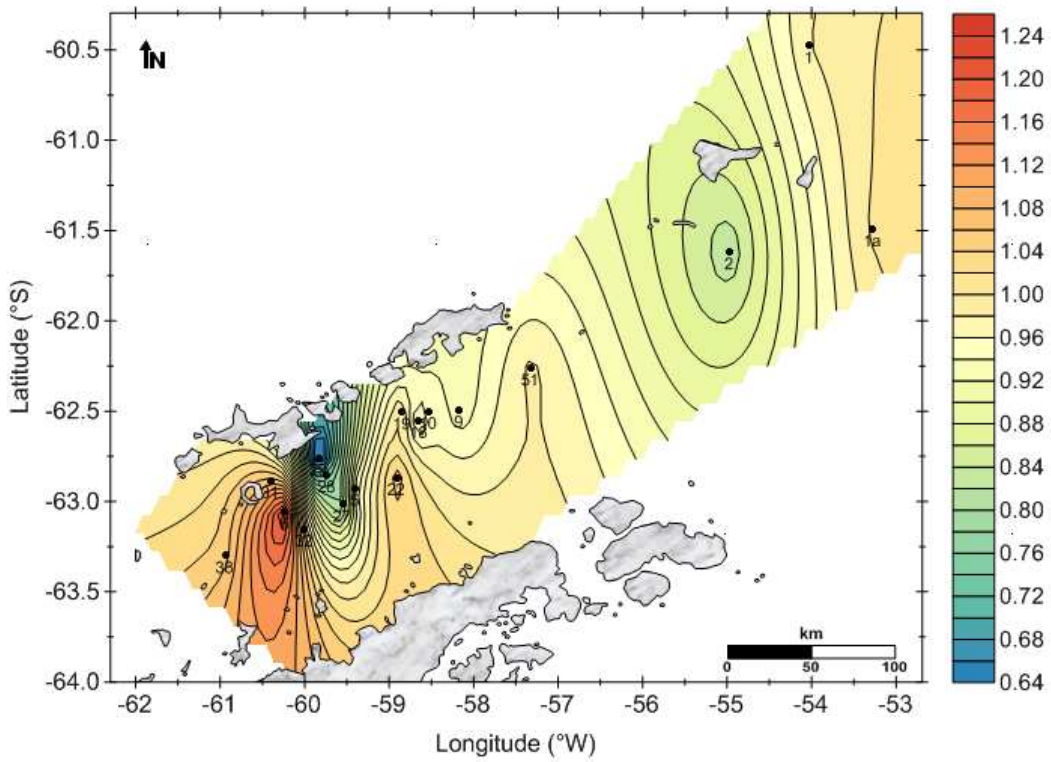

Figure 12: Total ^{234}Th/ ^{238}U AR distribution map of surface seawater collected at OPERANTAR XXX expedition.

Due to the fact of those 2 OPERANTAR expeditions were carried out in different temporal conditions, seasonal oscillations on total ^{234}Th and dissolved ^{238}U activities can be noticed. In October/2011 (beginning of summer season), ice melting processes were occurring intensively causing a major supply of total ^{234}Th compared to ^{238}U at North Bransfield Strait, possibly as a result of the presence of algae that accumulates ^{234}Th and release its excess to surface waters. These results can also indicate a combination of erosional continental input, freshwater and the formation of waters enriched in nutrients. In the case of total ^{234}Th, fluxes tended to be largest nearshore and decreased with increasing distance from land, showing the importance of the coast on ^{234}Th distributions. Taking into account all sampling stations established in March and October/11 the relative variability of total ^{234}Th distribution was 22%.

4. CONCLUSIONS

Research on the distribution of natural radionuclides in Antartica is rare and thus, there is a great interest in to know their occurrence and factors related to its mobilization, transference and accumulation in this extremely fragile environment. ^{234}Th is a particle-reactive radionuclide produced continuously in seawater by the decay of its soluble conservative precursor ^{238}U and the disequilibrium ^{234}Th/^{238}U in the surface ocean has been applied to estimate carbon fluxes exported in the Southern Ocean via sinking material. The flux of particles biologically productive out to the euphotic zone in the Southern Ocean has special attention due to its importance in the control of CO_2 atmospheric concentrations. Because this work focused on 2 OPERANTAR expeditions carried out during 2011 in different temporal

conditions, seasonal oscillations on total ^{234}Th and dissolved ^{238}U activities were verified. In October/2011 a major supply of total ^{234}Th compared to ^{238}U at North Bransfield Strait occured, indicating a combination of erosional continental input, freshwater and the formation of waters enriched in nutrients. Taking into account all sampling stations established in March and October/2011 the relative variability of total ^{234}Th distribution was 22%.

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