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Circulation of Circumpolar Deep Water and marine environment traced by ^{127}I and ^{129}I speciation in the Amundsen Sea Polynya, Antarctica

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Abstract: The long-lived anthropogenic ^{129}I released from human nuclear activities has been widely employed as an effective oceanographic tracer to investigate circulation of water masses in marine environment. Depth profiles of seawater collected from the Amundsen Sea Polynya, Antarctica were analyzed for total ^{129}I and ^{127}I, as well as their species of iodide and iodate. The measured ^{129}I concentrations ((1.15-3.43)×10^6 atoms/L) and $^{129}I/^{127}I$ atomic ratios ((0.53-1.19)×10^{-11}) indicate that anthropogenic ^{129}I has not only reached the Antarctic surface marine environment but also the deep water due to a strong vertical mixing of water masses. The Circumpolar Deep Water (CDW) flowed southward along continental shelf towards the ice shelf zone (74.25°S) at a depth of 1025 m and then migrated upward and northward to the polynya and finally to the sea ice zone (71.95°S). The maximum upwelling depth of the CDW was around 200 m in polynya. The source of $^{129}I^-$ in polynya is predominantly the intrusion of source waters rather than the in-situ reduction of iodate by phytoplankton, implying a considerably slow reduction process of iodate to iodide in this region.

Keywords: Iodine-129; iodine species; Circumpolar Deep Water; marine environment; Amundsen Sea Polynya

1. Introduction

A polynya is an area of open water surrounded by sea ice, where a relative higher biological production occur. The Amundsen Sea Polynya is one of the most productive regions of primary productivity in Antarctica (Arrigo et al., 2012; Arrigo et al., 2003), which reaches up to 2.2 g C m^{-2} day^{-1} during the austral spring and summer (Kim et al.,

2015; Lee et al., 2012). The growth and accumulation of phytoplankton biomass are mainly attributed to the relatively high seawater temperature, long solar radiation and abundant supply of nutrients from glacial melting and the intrusion of warm waters into the surface (Jacobs et al., 2011; Jenkins et al., 2010). During the formation of polynya, heat and nutrients from surface warm water migration or deep warm water upwelling can reduce sea ice cover located in waters that would be expected to be ice covered (Payne et al., 2007; Wåhlin et al., 2010; Walker et al., 2007) and promote primary productivity during the austral spring and summer (Grand et al., 2015; Hatta et al., 2016; Sedwick et al., 2011). Our previous research has found that the eastward flowing Antarctic Circumpolar Current (ACC) migrates southward into the Amundsen Sea through 69.4°S in the warm season (Xing et al., 2017b). The strong vertical exchange/mixture of Circumpolar Deep Water (CDW) between the surface and deep water in the Amundsen Sea was also observed (Xing et al., 2017a). However, the maximum upwelling layer and location of the CDW could not be identified in the Amundsen Sea Polynya because of the lack of more detailed data. Understanding the movement pathway of the CDW will provide critical information pertaining to the formation mechanism and ecosystem of the Amundsen Sea Polynya.

Due to its long residence time and high solubility in seawater, anthropogenic iodine-129 has been employed as an effective oceanographic tracer for the investigation of water movement in the marine environment (Hou et al., 2007; Hou et al., 2002; Keogh et al., 2007; Muramatsu et al., 2004; Xing et al., 2017b). Inorganic iodine in the seawater exists mainly as dissolved iodate and iodide. Iodide is a thermodynamically

unfavorable species in oxidized waters, but the reduction of iodate to iodide does not occur spontaneously. . A kinetic barrier inhibits the direct chemical oxidation of iodide to iodate (Wong 1991). The formation of iodide is generally controlled by biological processes including phytoplankton, bacteria and enzymes in the ocean, especially in coastal seas (Hou et al., 2007; Moisan et al., 1994; Tian et al., 1996; Wong 2002; Wong et al., 2001). Consequently, iodine species, in particular ^{129}I species, are a valuable oceanographic tracer for the investigation of the marine environment.

Beyond our previous research on total iodine isotopes, this work aims to investigate the vertical distribution of iodine species (^{129}I and ^{127}I) in seawater in the Amundsen Sea Polynya to better understand the movement of the CDW and its influence on the marine environment in this region. To achieve this purpose, total iodine isotopes (^{129}I and ^{127}I) and their species in five depth profiles were analyzed using chemical separation and sensitive measurement techniques of accelerator mass spectrometry (AMS) for ^{129}I and ICP-MS for ^{127}I.

2 Materials and methods

2.1. Samples and standards

Five seawater depth profiles were collected through a built-in seawater sampler in the research vessel N.B. Palmer during cruise in the Amundsen Sea Polynya, Antarctica (Fig. 1). Potentially suspended particles were removed by filtration through a 0.45 μm membrane filter, and the filtered seawater samples were stored in 2 L polyethylene bottles. The principal physicochemical parameters including seawater temperature,

89 salinity, chlorophyll a, dissolved oxygen, CO_2 partial pressure (pCO_2) and nutrients
90 (phosphate, nitrite, nitrate, ammonium, silicate) were determined on site. ^{129}I standard
91 solution (NIST-SRM-4949c) was supplied by the National Institute of Standards and
92 Technology (NIST, Gaithersburg, MD, U.S.A). ^{127}I carrier solution with low ^{129}I level
93 ($^{129}I/^{127}I$ atomic ratio < 2×10^{-14} carrier) was prepared by dissolution of Woodward
94 iodine crystals (Woodward Iodine Corporation, Oklahoma, U.S.A). All chemical
95 reagents were of analytical grade, and all solutions were prepared using deionized water
96 (18.2 MΩ·cm).

97 *2.2. Separation of iodine and its chemical species in seawater*

98 For the analysis of iodine-127 and its species, the original seawater sample and
99 1.0 mL of sample solution separated by AG 1-×4 anion exchange chromatography were
100 taken to a vial, and then diluted 15 times using 1% ammonium solution. ^{127}I in the
101 prepared samples containing 2 μg/L Cs^+ as the internal standard were measured using
102 an ICP-MS (X series II, Thermo Scientific, USA). The detection limit of the method
103 for ^{127}I was 0.02 μg/L. The iodide concentration was corrected by for its chemical yield
104 during chromatographic separation by means of a ^{125}I spike added.

105 About 0.6-1.2 L of seawater were employed for separation of iodine-129 and its
106 species. For separation of total iodine-129, 0.2 mg of ^{127}I carrier (as NaI, prepared from
107 Woodward iodine) and 1000 Bq of $^{125}IO_3^-$ spike were added to 0.6 L seawater, 0.5 mL
108 of 2 mol/L $NaHSO_3$ and 3 mol/L HNO_3 solution were added to convert different iodine
109 species to iodide. 28 mL of 0.01 mol/L $AgNO_3$ (30 mg Ag^+) was then dropwise added
110 to the solution under stirring to form an AgCl-AgI-AgBr-Ag_2SO_3 precipitate. The

precipitate was separated by centrifuge and sequentially washed with 6 mol/L HNO_3, H_2O, 35% and 25% ammonium to remove Ag_2SO_3, most of the AgCl and AgBr until a final precipitate of 1-3 mg was acquired. For separation of iodine species, 1000 Bq of $^{125}I^-$ spike and 0.2 mg of $^{127}I^-$ carrier ($^{129}I/^{127}I$ atomic ratio < 2.0×10^{-13}) were added to 1.2 L seawater, and then $NaHSO_3$ was added to final concentration of 0.3 mmol/L. 0.5 mol/L HNO_3 was slowly added into the solution under stirring to adjust pH ~5. 45 mL of 0.03 mol/L $AgNO_3$ (150 mg Ag^+) was dropwise added under stirring to form an AgCl-AgI-AgBr-Ag_2SO_3 precipitate. The precipitate was separated by centrifuge and the supernatant was employed for separation of iodate. The following procedure was the same as those for total iodine. The detailed analytical method has been reported elsewhere (Luo et al., 2013; Xing et al., 2017a). The schematic diagram of the analytical procedure is shown in Figure S1 in the Supplementary materials. ^{125}I was measured using a NaI gamma detector, and chemical yields for iodine and its species in the whole procedure were higher than 80%. Procedural blanks were prepared using the same procedure mentioned above but no samples were added.

Two ^{129}I standard solutions, with a total iodine concentration of 1 mg/ mL and $^{129}I/^{127}I$ atomic ratios of 9.954×10^{-12} and 1.138×10^{-10} respectively, were prepared by dilution of ^{129}I standard solution with ^{127}I carrier solution. Two ^{129}I working solutions were prepared by mixing the above prepared ^{129}I standard solution with NaCl solution in Cl/I mass ratio of 2:1. The AgI-AgCl precipitate was dried at 60-70 °C and mixed with niobium powder in a mass ratio of 1:5, which was finally pressed into copper holders using a pneumatic press for AMS measurement of ^{129}I in the Xi'an AMS center.

3 Results

3.1. Physicochemical parameters

The results of salinity, temperature, chlorophyll a, pCO_2, dissolved oxygen and nutrients, including nitrite, ammonium, phosphate, nitrate and silicate, in the Amundsen Sea are given in Table 1. According to the vertical distribution of salinity and temperature, the seawater was divided into three layers, including 1) Antarctic Summer Surface Water (SSW), 2) Antarctic Low Salinity Shelf water (LSSW), 3) Circumpolar Deep Water (CDW). The chlorophyll a concentrations in the surface water of the profile-2 and profile-3 were 21.5 μg/L and 5.68 μg/L, respectively, which were significantly higher than values reported for most of the Southern Ocean (<0.5 μg/L) (Fukuchi 1980), suggesting a very active phytoplankton population. The pCO_2 in surface seawater of the profile-2, profile-3, profile-4 in polynya ranged from 196.0 μatm to 247.5 μatm, which were remarkably lower than that of the profile-1 in the sea ice zone (404.9 μatm) and profile-5 in the ice shelf zone (447.5 μatm). On the contrary, the concentrations of dissolved oxygen in surface seawater in polynya (8.52-9.91 mg/L) were higher than that in the sea ice zone (7.00 mg/L) and the ice shelf zone (7.41 mg/L). The vertical variation of dissolved oxygen in all profiles showed a decrease trend with depth. The concentrations of dissolved oxygen ranged from 6.27 mg/L to 7.03 mg/L at depths of 100-300 m and from 4.24 mg/L to 4.79 mg/L at depths ＞300 m. The distribution patterns of nutrients in the surface seawater showed a lower concentration in the polynya as compared to the sea ice zone and the ice shelf zone. The vertical

variations of nutrients increase with depth and the lowest concentrations lay near the surface (<100 m).

3.2. Vertical distribution of ^{129}I and ^{127}I species in seawater

The ^{129}I concentrations and $^{129}I/^{127}I$ atomic ratios in five seawater profiles and iodide/iodate molar ratios in profile-3 and profile-5 are presented in Fig. 2, Table S1 and Table S2 in Supplementary. The ^{129}I concentrations ranged from 1.54×10^{6} atoms/L to 3.43×10^{6} atoms/L (Fig. 2a), which were more than one order of magnitude lower than what was found in the Northern Hemisphere (> 1.0×10^{7} atoms/L) (Snyder et al., 2010). The $^{129}I/^{127}I$ atomic ratios ranged from 0.53×10^{-11} to 1.19×10^{-11} (Fig. 2b) with an average of 0.81×10^{-11}, which were 5.4 times higher than the pre-nuclear level (1.5×10^{-12}) in the marine system (Fehn et al., 2000; Moran et al., 1998), indicating that anthropogenic ^{129}I has not only reached the Antarctic surface water but also the deep water with a depth of at least 1025 m. The vertical variations of ^{129}I concentrations and $^{129}I/^{127}I$ atomic ratios in this region fluctuated with depth, indicating a strong vertical exchange/mixture of water masses between the surface and deep water. The highest values of ^{129}I concentration and $^{129}I/^{127}I$ atomic ratio in profile-1, profile-3 and profile-4 lay at depths of 200-500 m while in profile-5 lay at a depth of 800 m. Compared with ^{127}I species in profile-3 and profile-5 (Fig. 2c), the iodide/iodate molar ratios of ^{129}I showed significantly different patterns. The $^{127}I^{-}/^{127}IO_3^{-}$ molecular ratios ranged in 0.06-0.25, the $^{129}I^{-}/^{129}IO_3^{-}$ molecular ratios increased from 1.50 in the surface to 5.58 in the deep water, indicating that ^{129}I exists predominantly as iodide, while ^{127}I as iodate.

4 Discussion

4.1. Level and vertical distribution of total ^{129}I in the Antarctic

Despite plenty of ^{129}I depth profiles in seawater from many locations are available, seven representative profiles spread in different oceans were selected for comparison in this work (Fig. 3) (Alfimov et al., 2004; Hou et al., 2013; Povinec et al., 2011; Schink et al., 1995; Smith et al., 1998; Suzuki et al., 2008). It can be seen that the lowest ^{129}I concentrations in the surface seawater occur in the Antarctic ((0.016-0.030)×10^8 atoms/L). The high values ((3.50-15.53)×10^8 atoms/L) were observed in the Amundsen Basin, Makarov Basin and Beaufort Sea in the Arctic (Alfimov et al., 2004; Chang et al., 2019; Smith et al., 1998). They were 10^2-10^3 times higher than those observed in the Antarctic. This results from the transport of huge amount of marine discharged ^{129}I from the nuclear fuel reprocessing plants (NFRPs) at La Hague (France) and Sellafield (U.K.) to the Arctic. ^{129}I concentrations in seawater collected from the mid-latitude Northern Hemisphere were (0.23-1.28)×10^8 atoms/L in offshore Fukushima (Hou et al., 2013) and Toyama Bay (Suzuki et al., 2008), and 0.15×10^8 atoms/L at the Gulf of Mexico (Schink et al., 1995). These values were approximately two orders of magnitude higher than those we observed in the Antarctic. Only a few ^{129}I data from the Southern Hemisphere were available, where even the considerably high ^{129}I concentration (0.08×10^8 atoms/L) in seawater collected from Crozet Basin in the South Indian Ocean (Povinec et al., 2011) was approximately 3-5 times as high as those in the Antarctic.

The vertical variation of ^{129}I concentrations in Antarctica shows a complex fluctuation in the whole profile (Fig. 3). The relatively high ^{129}I values occur at depths of 200 m, 500 m and 800 m, indicating strong vertical mixing of water masses between the surface and deeper layers. Similarly, the variation of ^{129}I concentrations in profile from the Crozet Basin in Indian Ocean shows the highest value (1.5×10^{7} atoms/L) around the 200 m and drops to 3.3×10^{6} atoms/L at a depth of 1000 m and then fluctuates between 3.4×10^{6} and 7.7×10^{6} atoms/L down to 5000 m (Povinec et al., 2011). It indicates that the intrusion of different water masses such as the North Indian Deep Water (NIDW) and the North Atlantic Deep Water (NADW) occurred in this area. However, depth profiles of ^{129}I in Amundsen Basin, Makarov Basin, Fukushima and Gulf of Mexico have the highest ^{129}I concentrations in the surface (100 m), which are at least 2-5 times as high as those in the subsurface (200 m), and then decrease exponentially with depth (Alfimov et al., 2004; Hou et al., 2013; Schink et al., 1995), indicating a dynamic horizontal movement of water mass in the surface seawater. The low ^{129}I concentration of seawater collected from the Gulf of Mexico in 1992 was $(3\pm2)\times10^{5}$ atoms/L at a depth of 1500 m, which was close to the pre-nuclear level (3×10^{5} atoms/L) (Snyder et al., 2010). The high ^{129}I concentrations of depth profiles in the Beaufort Sea (5.9×10^{9} atoms/L) and Toyama Bay (2.6×10^{6} atoms/L) lay in the subsurface layer (~ 200 m), followed by a sharp decrease with depth (Smith et al., 1998; Suzuki et al., 2008). The ^{129}I distribution pattern may reflect the intrusion of different water masses, as observed in the Beaufort Sea, with water from Pacific Ocean with relatively low ^{129}I, and the subsurface water that contains elevated ^{129}I from European

NFRPs.

4.2. Transport pathways of CDW in the Amundsen Sea Polynya

In order to explore water circulation using anthropogenic ^{129}I, the major source of ^{129}I in the Antarctica marine water has to be identified. The investigations have shown that anthropogenic radionuclides including ^{129}I in the Antarctica marine water originated from fallout of the nuclear weapons testing in 1945-1980, especially those conducted in the Pacific Proving Grounds (low latitude area of the North Pacific Ocean) in the 1950s. The large releases of ^{129}I from European nuclear reprocessing plants could not reach to this region (Xing et al. 2017b; Xing 2015). Distribution of ^{129}I in seawater has been successfully applied to explore movement of water masses in many areas (Bautista et al., 2016; Chang et al., 2016; Hou et al., 2007; Xing et al., 2017b; Yi et al., 2012). Fig. 4 shows the depth distributions of ^{127}I, ^{129}I, $^{129}I/^{127}I$, salinity and temperature in seawater in the Amundsen Sea Polynya. The ^{127}I concentrations in all depth profiles ranged from 0.44 μM to 0.50 μM (with an average of 0.48 μM), showing a fairly small variation. No correlation between ^{127}I concentrations and 129I/^{127}I atomic ratios (R^2=0.005) was observed. While, a remarkably positive correlation between ^{129}I concentrations and $^{129}I/^{127}I$ atomic ratios (R^2=0.998, $p<0.01$) in the investigated area was obtained. The ^{129}I concentrations in surface seawater in the profile 2-5 ranged from1.55×10^6 to3.08×10^6 atoms/L, with an average of2.58×10^6 atoms/L, which mainly contributed to the southward migration branch of the ACC that carried the relatively high concentration of ^{129}I (2.75×10^6 atoms/L) into this region through location 6 (69.4°S) (Xing et al., 2017b). The ^{129}I concentration in surface seawater in the profile-

1was (1.98±0.12)×10^6 atoms/L, which were lower than that in other profiles, suggesting that the movement of the ACC branch in this region may be restricted due to sea ice coverage.

The ^{129}I concentrations in CDW obviously decreased with the distance away from the ice shelf zone ((3.43±0.64)×10^6 atoms/L at a depth of 800 m) to the polynya ((2.81-3.1)×10^6 atoms/L at depths of 200-440 m) and then to the sea ice zone ((3.02±0.16)×10^6 atoms/L at a depth of 250 m). Consequently, the pattern of ^{129}I distribution in CDW demonstrated a major transport pathway of waste masses from the ice shelf zone upwelling northward to the polynya and finally to the sea ice zone (71.95°S), and the further migration southward of CDW can reach to the continental shelf (74.25°S) in the Amundsen Sea, which was similar to the transport pathway of CDW that can reach to continental shelf (67.5°S) in the Prydz Bay (Yabuki et al., 2006). The CDW was able to rise up to the depth of up to around 200 m in the polynya.

4.3. Sources of iodide in the Amundsen Sea Polynya

As shown in Table 1, the pCO_2 and the nutrient concentrations in the surface water in the profile-3 in the polynya were remarkably lower than those in profile-5 in the ice shelf zone. In contrast, the concentration of dissolved oxygen in the surface water (9.91 mg/L) in the profile-3 was higher than in the profile-5 (7.41 mg/L). These results suggest that the phytoplankton activity in the surface water in polynya was much more vigorous than in the ice shelf zone. The iodide/iodate molar ratios of ^{127}I and ^{129}I in profile-3 and profile-5 show that ^{127}I and ^{129}I exist mainly as iodate and iodide,

respectively (Fig. 2c). It is well acknowledged that biological activity can convert iodine species especially in coastal areas (Hou et al., 2007). The molecular ratio of 0.17 for $^{127}I^-/^{127}IO_3^-$ and 1.5 for $^{129}I^-/^{129}IO_3^-$ were observed in depth of 2 m in the polynya, which was lower than these of 0.25 and 2.38 in depth of 200 m. The difference of molecular ratios of iodide/iodate between the surface and subsurface seawater suggest that part of the iodate might be converted to iodide in the seawater with very active phytoplankton population (Wong 2002), and the reduction of iodate to iodide in the polynya might be a relatively slow process.

As discussion above, a branch of ACC with the high ^{129}I concentration (2.75×10^6 atoms/L) driven by the radial wind stress moved southward across the sampling location 6 and reached the investigated area. The $^{129}I^-/^{129}IO_3^-$ molecular ratio at location 6 (1.17) was slightly lower than that in the polynya. $^{129}I^-$ in the surface water in the polynya, therefore, was mainly controlled by the contribution of branch of ACC. Due to the strong vertical mixing of water masses, the high $^{129}I^-/^{129}IO_3^-$ values in the deeper layer originated from the upwelling of CDW carried the high $^{129}I^-/^{129}IO_3^-$ values. These observations confirm that the analysis of ^{129}I species can be very helpful to investigate the mixing and circulation of water masses.

5 Conclusions

This study reported the level and vertical distribution of total ^{129}I and ^{127}I and their species (iodide and iodate) in the Amundsen Sea Polynya, Antarctica. ^{129}I concentrations of $(1.15\text{-}3.43)\times10^6$ atoms/L and $^{129}I/^{127}I$ atomic ratios of $(0.53\text{-}1.19)\times10^-$

^{11} are the lowest compared with the other oceans/seas, but still about one order of magnitude higher than the pre-nuclear level, indicating that anthropogenic source of ^{129}I in the Antarctic marine environment has reached the Antarctic deep water down to 1025 m. The distribution pattern of ^{129}I concentrations in CDW, which decreased from the ice shelf zone of $(3.43\pm0.64)\times10^6$ atoms/L to the Amundsen Sea Polynya of $(2.81-3.1)\times10^6$ atoms/L and the sea ice zone of $(3.02\pm0.16)\times10^6$ atoms/L, demonstrated a major transport pathway from the ice shelf zone upwelling northward to the sea ice zone (71.95°S). The high $^{129}I^-/^{129}IO_3^-$ molar ratios in seawater should mainly be attributed to the intrusion of source waters — a branch of ACC and CDW — carried the high $^{129}I^-/^{129}IO_3^-$ values. $^{129}I^-$ in the polynya was also controlled by the input of ACC.

Declaration of interest

There are no conflicts to declare.

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Table 1 Results of physicochemical parameters in the Amundsen Sea

Caption of figures

Fig. 1 Sampling locations (indicated as blue dots) of depth profiles in the Amundsen Sea Polynya, Antarctica. Sea ice zone, Amundsen Sea Polynya, and ice shelf zone are indicated by black circle, yellow circle, and red circle, respectively.

Fig. 2 Depth distributions of ^{129}I concentration (a), $^{129}I/^{127}I$ atom ratio (b) and iodine species ratio (c) in depth profiles.

Fig. 3 Comparison of depth distribution of ^{129}I concentrations in seawater in the Antarctic and other oceans.

Fig. 4 Vertical distributions of ^{127}I, ^{129}I, $^{129}I/^{127}I$, salinity and temperature in the Amundsen Sea Polynya.