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Tracing of water masses using a multi isotope approach in the southern Indian Ocean

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ABSTRACT

Anthropogenic radionuclides (^{3}H, ^{14}C, and ^{129}I) stemmed from nuclear weapons tests were found in 1999 to be very abundant in the surface of the southern Indian Ocean, comparable to those in the subtropical Northwest Pacific Ocean. The observed radionuclide variations with latitude/longitude in the southern Indian Ocean are not due to deposition patterns of global fallout, but due to transport of water masses from the western Pacific through the Indonesian seas, and different water fronts present in the Crozet Basin of the Indian Ocean. High radionuclide concentrations observed in the latitudinal belt of 20-40°S are associated with the Indian Ocean Subtropical Gyre which acts as a reservoir of radionuclides, maintaining their high concentrations on a time scale of several decades. ^{14}C data documents that the southern Indian Ocean is an important for sink of anthropogenic carbon. The isotopic tracers reveal the evidence of the most intense surface gradients and presence of several water masses in the southern Indian Ocean, which makes the region one of the most dynamic places of the World Ocean.

1. Introduction

Global fallout radionuclides (e.g. tritium (^{3}H), radiocarbon (^{14}C), strontium-90 (^{90}Sr), cesium-137 (^{137}Cs), iodine-129 (^{129}I), americium-241 (^{241}Am), plutonium isotopes (^{238}Pu, ^{239,240}Pu), etc.,) have been found as useful tracers for studying the heat and material transport and exchange processes occurring naturally both in the terrestrial (e.g. Hou et al., 2009; Levin and Hesshaimer, 2000; Santschi and Schwehr, 2004) and marine environments (e.g. Livingston and Povinec, 2002; Schlosser et al., 1999). Concentrations of these radionuclides in the sea had risen since 1945 and peaked in the Northern Hemisphere in 1963, after large scale atmospheric nuclear weapons tests carried out in 1961-1962 by former Soviet Union at Novaya Zemlya (Livingston and Povinec, 2002). In the equatorial Pacific close in fallout from nuclear weapons tests carried out at Bikini and Enewetak Atolls contributed to radionuclide inventories in the Pacific Ocean as well. The major portion of global fallout deposited in the mid-latitudes of the Northern Hemisphere (UNSCEAR, 2000), in particular, the North-western Pacific due to the combined effect of higher precipitation and higher stratosphere-troposphere exchange of air (Aoyama et al., 2006). Some of the global fallout radionuclides (e.g. ^{3}H, ^{14}C, ^{90}Sr, ^{137}Cs, ^{129}I) are dissolved in seawater and becomes constituents of seawater, and suitable therefore for studying transport of water masses in the ocean. On the other hand Am and Pu isotopes are more particle reactive (La Rosa et al., 2005), and suitable for investigation of processes in the water column and sediments. Significant portions of these radionuclides in the world ocean have accumulated at the seafloor as bottom sediments (Bowen et al., 1980; Hong et al., 1999; Lee et al., 2005; Livingston et al., 2001).

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We shall focus in this paper on three radionuclides, ^{3}H, ^{14}C and ^{129}I. Tritium is directly incorporated into the water molecule, usually as HTO, and has suitable half-life (12.32 y), therefore, it is used extensively in oceanographic studies. It is produced both naturally by interactions of cosmic rays with nitrogen and oxygen in the upper atmosphere, and artificially in large amounts from atmospheric nuclear weapons tests. Its concentration peaked in the atmospheric moisture of the Northern Hemisphere in 1963, when it was 1000 times higher than its cosmogenic concentration of 60 TU (Weiss and Roether, 1980). It has also been released in large quantities from nuclear reprocessing facilities (Livingston and Povinec, 2000). The penetration of bomb tritium from surface waters into deeper layers of the ocean has been used to study pathways and time scales of deep and bottom water formation, (e.g. Bayer and Schlosser, 1991; Broecker and Peng, 1982; Broecker et al., 1986). Tritium with its strong interhemispheric concentration gradient (compared e.g. with radiocarbon) is a unique tracer for studying exchange of water masses between the basins.

Natural ^{14}C levels in the environment have also been disturbed by bomb ^{14}C. In 1963 they were in the carbon dioxide of the Northern Hemisphere by a factor of 2 higher than natural levels (Burchuladze et al., 1989; Nydal and Lövseth, 1965). Because of its long half-life (5730 y) and subsequent formation of $^{14}CO_2$ in the air, specific stratosphere-troposphere-biosphere mixing, exchange of carbon dioxide with surface ocean, and sequestration of carbon dioxide into the deep ocean, ^{14}C became the most frequently studied environmental radionuclide, important for better understanding of climate change (e.g. Key et al., 2004). Oceanic radiocarbon data contributed to a better understanding of thermohaline circulation in the World Ocean known as a Great Ocean Conveyor Belt (Broecker, 1991; Rahmstorf, 2006).

^{129}I has been introduced to the World Ocean from nuclear reprocessing facilities, global fallout and due to its natural production (Raisbeck and You, 1999). Because of its pulsed input from global fallout, releases by reprocessing plants in northwestern Europe, and long-half-life (15.7 million years) it is a powerful tracer to delineate source waters in the North Atlantic (Edmonds et al., 2001).

Thanks to recent developments in mass spectrometric analysis (^{3}He ingrowth from tritium decay; Clarke et al., 1976; Schlosser et al., 1999; Top, 1999), and accelerator mass spectrometry for ^{14}C (e.g. Jull et al., 2008; Key et al., 1996;2002; Povinec, 2005; Povinec et al., 2008; Tuniz et al., 1998) and ^{129}I (e.g. Kilius et al., 1987; Povinec et al., 2000; Raisbeck and You, 1999) it has been possible to analyze these radionuclides with high sensitivity and precision in small seawater volumes (e.g. 0.5 L), and thus investigate their distribution in the water column using direct sampling with Rosette systems. Also, multinational oceanographic sampling program has been emerged to cover the synoptic view of the global ocean. A notable one is the WOCE (World Ocean Circulation Experiment) program conducted in the 1990 s, which represents the most extensive ^{3}H and ^{14}C project carried out in the World Ocean (Key, 1996; www.eWOCE.org).

Although global fallout radionuclide distribution datasets have been frequently used as tools for tracing water masses in the World Ocean (e.g. Livingston and Povinec, 2002; Schlosser et al., 1999;2001), the Indian Ocean has received only a limited attention (Lee et al., 2009; Povinec et al., 2004a; van Beek et al., 2008). The first measurements of ^{3}H and ^{14}C were made through the Geochemical Ocean Sections (GEOSECS) project (1977-1978) (Broecker et al., 1986; Östlund and Brescher, 1982; Stuiver and Ostlund, 1983), and followed by the WOCE field project (1990-1998; www.eWOCE.org) in the Indian Ocean. Unfortunately, the ^{3}H and ^{14}C measurements were not carried out in the western Indian Ocean south of 34°S.

The Indian sector of the Southern Ocean is a key region for the exchange of water masses between Antarctica and Equatorial regions, playing an important role in the global climate change (Key et al., 2004). The Southern Ocean is the largest oceanic high-nitrate low chlorophyll region in the world. It is contributing to the regulation of the atmospheric CO_2 via the biological pump (Metzl et al., 1999). The southern Indian Ocean was included therefore in the Worldwide Marine Radioactivity Studies (WOMARS), coordinated by the International Atomic Energy Agency's Marine Environment Laboratories (IAEA-MEL) in Monaco, and carried out in collaboration with several laboratories in Denmark, France, Germany, India, Italy, Japan, South Korea, New Zealand, Sweden, UK and USA (International Atomic Energy, 2005; Povinec et al., 2003a; Povinec et al., 2005). The aim of the project was to study the distribution and behavior of anthropogenic radionuclides in the world ocean. Some of the results obtained for the northern Indian Ocean have already been published (Bhushan et al., 2003; Mulsow et al., 2003; Povinec et al., 2003b). French ANTArctic RESearch (ANTARES) IV cruise in the southern Indian Ocean in 1999 was liaised with the IAEA WOMARS program. The cruise plan (Park et al., 2002) and strategy for sampling radionuclides (Coppola et al., 2005, 2006; Lee et al., 2009) was designed with the aim to study distribution of radioactive and stable isotopes in the Crozet Basin of the southern Indian Ocean characterized with strong ocean currents.

In this paper, we report the distribution of radioactive (^{3}H, ^{14}C and ^{129}I) and stable (^{2}H, ^{18}O) isotopes in surface and deep waters of the southern Indian Ocean. Results on ^{90}Sr ^{239,240}Pu and ^{241}Am in surface waters and plankton have been published earlier (Lee et al., 2009).

2. Oceanography background

The Indian Ocean is limited northward to 25°N by the continent, and at 60°S, where it becomes the Southern Ocean. The 60°S limit appears to be largely set by political grounds. An important feature of water circulation in the Indian Ocean is a transport of warm water masses (10 - 15 Sv) from the western Pacific Ocean via the Indonesian throughflow to the eastern Equatorial Indian Ocean and to the southern Indian Ocean (Fine, 1985; Gordon and Fine, 1996; Gordon et al., 2003). The south Indian Ocean Subtropical Gyre (IOSG) is the most important current system influencing water circulation between 20° and 40°S (Tomczak and Godfrey, 1994). The gyre is the most intense with a tight, double celled central core focused at the western boundary. A fraction of IOSG water is transported southward by the Agulhas Current (AC) along the eastern African coast, entering the South Atlantic around the Cape of Good Hope, forming a branch of the AC that does not complete the retroflection pattern, also called the Agulhas leakage (Gordon, 1985; Schmitz, 1995).

Along the eastern boundary of the Indian Ocean, off the western Australia, the Leeuwin Current flows poleward along the continental shelf break from about 22°S to 35°S, and then turns eastward. The Leeuwin Current is warm and of relatively low salinity, low dissolved oxygen and high phosphate content. It transfers a significant amount of heat to the south (Tomczak and Godfrey, 1994).

South of the 40°S, the wind-driven Antarctic Circumpolar Current (ACC), going around Antarctica, transports cold waters from the Atlantic Sector of the Southern Ocean via the southern Indian Ocean to the southern Pacific Ocean. The southern Indian Ocean plays therefore a key role in the exchange of water masses between the Equator and Antarctica (Ganachaud and Wunsch, 2000), and between the Southern Hemisphere basins (Ridgway and Dunn, 2007), important for better understanding of global oceanic processes and the climate (Key et al., 2004; Rahmstorf, 2002). As it is surrounded by highly populated continents in its Northern area, there is also subjected to contamination from land-based sources.

The banded structure of the ACC in the southern Indian Ocean consists of several narrow jets associated with sharp hydrographic fronts due to the presence of the Crozet and Kerguelen Plateaus (Park et al., 1993) (Fig. 1). From the north of the Crozet Islands to the downstream area, there exists a very strong triple frontal zone where