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# Mercury and methylmercury in the Atlantic sector of the Southern Ocean



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## ABSTRACT

Oceans constitute one of the most important reservoirs for mercury. In order to provide a first insight into the concentrations of Hg species in the Atlantic sector of the Southern Ocean a sampling campaign was carried out south of the Polar Front.

Water samples taken at discrete depths from the surface down to 300 m at six stations were analysed for total Hg (HgT), methylmercury (MeHg) and other interpretative parameters such as salinity, temperature, dissolved and particulate organic carbon, dissolved oxygen, chlorophyll and inorganic nutrients.

Results showed a high spatial variability in the concentrations of HgT and MeHg. HgT ( $0.93 \pm 0.69 \text{ ng L}^{-1}$ ) and MeHg ( $0.26 \pm 0.12 \text{ ng L}^{-1}$ ) levels were similar or higher than those reported in previous works in high latitude studies. The highest values were found at a location ( $-53^\circ, 10^\circ\text{E}$ ) south of the South Polar Front, an area of strong gradients caused by the mixing of different water masses. Vertical profiles showed a great variability even for those stations sampled at the same location or an area dominated by the same oceanographic features. A decrease of HgT and a consequent increase in MeHg with depth was observed in some sites, suggesting the occurrence of Hg-methylation process, while at other stations, a concurrent decrease or increase of both mercury species was observed. In spite of these differences, an overall positive correlation between HgT and MeHg was observed. Differences between vertical profiles of Hg species were attributed to favourable environmental conditions for Hg methylation. The highest proportion of MeHg (% of HgT) was observed in sites with low dissolved oxygen or highest estimated remineralization rates.

The results obtained in this study show that the Hg distribution and speciation in the Atlantic sector of the SO is comparable (or in some sites higher) to the ones published for the other open ocean regions. However, the concentrations of MeHg in this area are more dependent on the environmental conditions than on the total concentration of Hg present in the water.

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## 1. Introduction

Mercury (Hg) is a globally distributed pollutant that cycles between air, water, sediment, soil and organisms (Moreno et al., 2005) and is considered by the United Nations Environmental Programme (UNEP) as a pollutant of great concern. Its presence in the environment is increased due to anthropogenic sources, and can create fatal

health issues. Although anthropogenic emissions of Hg have been reduced by half in the last decades (Pacyna et al., 2001), ongoing contamination is still a worldwide problem. The conversion of inorganic Hg into methylmercury (MeHg;  $\text{CH}_3\text{-Hg}^+$ ), a strong neurotoxin, is a critical step in its fate and toxicity. Interest in MeHg as an environmental contaminant first arose in the 1960s with reports of alkylmercury poisoning of marine life and humans in Japan, and birds and marine life in Sweden (Kiyoura, 1964; Johnels and Westermark, 1969). The net MeHg concentrations result from the balance between methylation and demethylation processes, which are not completely understood (Mason and Benoit, 2003). The most important factors

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influencing biological methylation are the availability of inorganic mercury and the nature of the microbial community (Mauro et al., 1999; Ullrich et al., 2001) modulated by physical and chemical parameters such as temperature, pH, salinity, organic carbon, and redox potential (Gilmour and Henry, 1991; Barkay et al., 1997; Ullrich et al., 2001; Mason and Benoit, 2003). Despite the paucity of information concerning sources, in situ production, biogeochemistry and bioaccumulation of MeHg in marine organisms, sediments appear as potentially significant sources of MeHg to food webs in the coastal zone (Mason et al., 1999; Gill et al., 1999; Covelli et al., 1999; Langer et al., 2001; Hammerschmidt et al., 2004) and to the open ocean via hydrological or biological transport.

The ocean plays a central role in global mercury biogeochemical cycling. Within the ocean, Hg cycles among elemental ( $\text{Hg}^0$ ), inorganic ( $\text{Hg}^{2+}$ ), MeHg, dimethylmercury (DMHg), and particle-bound ( $\text{Hg}_p$ ) forms (Mason and Fitzgerald, 1993). Atmospheric wet and dry depositions supply Hg to the surface ocean. In the mixed layer, Hg(II) may be (photo)reduced to  $\text{Hg}^0$  and then re-emitted to the atmosphere (Qureshi et al., 2009). Hg(II) can also be adsorbed onto suspended organic-rich particulate matter ( $\text{Hg}_p$ ). Deeper ocean remineralization of sinking particulate matter converts  $\text{Hg}_p$  to dissolved Hg(II), thus resupplying Hg to the dissolved pool at depth (Mason et al., 1995; Amyot et al., 1997; Rolfhus and Fitzgerald, 2001; Fitzgerald et al., 2007; Strode et al., 2007). The hypoxic (low-oxygen) zone in the subsurface ocean is hypothesised to be an area of optimum biological methylation/demethylation processes (Sunderl and et al., 2009; Lehnher et al., 2011).

Burial of  $\text{Hg}_p$  in marine sediments is the terminal sink for Hg (Lamborg et al., 2003). Vertical profiles in ocean waters generally contain the signatures of the following three different processes:

- (i) In the mixed layer, total mercury ( $\text{Hg}_T$ ) concentrations can exhibit a maximum or minimum relative to subsurface waters depending on the relative roles of sources (atmospheric deposition and upwelling of Hg-rich waters) and sinks (air–sea gas exchange and scavenging onto particles) (e.g., Zhang et al., 2014);
- (ii) The thermocline/intermediate waters (between the bottom of the mixed layer down to 1000 m depth) often display a local maximum in  $\text{Hg}_T$  (Zhang et al., 2014). This maximum has been interpreted as a Hg release from remineralization of sinking particulate matter. This maximum can also be interpreted as an anthropogenic signal from high-deposition regions through isopycnal transport or adsorption onto sinking particulate matter (Mason et al., 1998; Mason and Sullivan, 1999; Fitzgerald et al., 2007; Strode et al., 2007).
- (iii) In the deep ocean (> 1000 m),  $\text{Hg}_T$  concentrations are relatively constant and sometimes slightly increase with depth (e.g., Laurier et al., 2004; Lamborg et al., 2012).

Many studies have been published about the Hg cycling in the world oceans (see references above) although, and with the exception of the biological compartments, less attention has been given to the Southern Ocean (SO). The first paper aiming to better understand the “abiotic” mercury cycle in the Southern Ocean was published by Cossa et al. (2011). In a scientific cruise between Tasmania and Antarctica, Cossa et al. (2011) found that  $\text{Hg}_T$  concentrations in water samples were comparable to recent measurements made in other parts of the world's oceans, however the Hg species distribution suggested distinct features in the SO Hg cycle: a net atmospheric Hg deposition on surface water near the ice edge, an Hg enrichment in brine during sea ice formation, and a net methylation of Hg.

The work presented here contributes to the scientific efforts to understand the Hg cycling in the SO. During a RV Polarstern cruise in the Atlantic sector of the SO, several water profiles down to

300 m were sampled and analysed for total Hg, MeHg and other interpretative parameters (e.g., salinity, temperature, dissolved and particulate organic carbon, etc). The aim of this work is, therefore, to provide a first insight into the concentrations of Hg species in this area and to understand the processes responsible for their distribution, partitioning and speciation.

## 2. Material and methods

### 2.1. Sampling

Sampling was carried out as part of the RV Polarstern “Eddy–Pump – ANTXXVIII/3” cruise along a longitudinal transect in the Atlantic sector of the Southern Ocean around latitude 50°S between January and March 2012 (Fig. 1).

Four different areas were sampled for Hg from east to west (see Table 1 for a more detailed position): in the east station 84 was located south of the Southern Polar Front (SPF) at 10°E, two stations were located in the central Atlantic sector (stations 91 and 140) between the SPF and the Antarctic Polar Front (APF) where a large open ocean diatom bloom was investigated (up to  $2 \mu\text{g chl-a L}^{-1}$ ), the third region was further to the west (stations 163 and 174) between the APF and SPF at the North end of the South Georgia basin. Finally, station 143 was located close to South Georgia in the SPF (receiving iron inputs from the vicinity of the island, data not published). Details of the addressing fronts, their locations and biogeochemical features are provided in this issue (Strass et al., 2017; Hoppe et al., 2017; Puigcorb  et al., 2017). The first station (st. 84) was characterised by low plankton biomass ( $< 0.5 \mu\text{g chl-a L}^{-1}$ ) in the euphotic layer (caused by a bloom in its declining stage; Cheah et al., 2017) and the surge of high iron/low oxygen waters of the Upper Circumpolar Deep Water (UCDW) which effect can be tracked up to 150 m deep (Puigcorb  et al., 2017). The second and third areas presented different development stages of a phytoplankton bloom. The last area, close to South Georgia, provides information on the influence of the “Island effect” on the levels and biogeochemical cycle of Hg in these waters.

Profiles of temperature, salinity and pressure were measured with a Seabird SBE 911plus CTD (conductivity–temperature–density; Strass et al., 2017). Samples for chlorophyll a, DOC, POC and macronutrients (nitrate+nitrite, phosphate, silicic acid) analyses were collected from Niskin bottles attached to the CTD.

Since solar radiation may change and reduce some forms of mercury, direct solar radiation data have been included in Table 1 for each station during the sampling time (LI-COR 192SA light sensor). Half of the stations (84, 143 and 163) were sampled at night, while the rest of the stations showed a radiation range between 34 and  $795 \text{ W m}^{-2}$ .

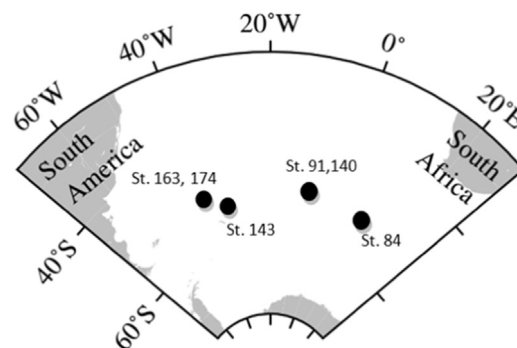


Fig. 1. Study area with the sampling stations.

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