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# Physical speciation of iron in the Atlantic sector of the Southern Ocean along a transect from the subtropical domain to the Weddell Sea Gyre

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[1] Distributions of total dissolvable iron (TDFe; unfiltered), dissolved iron (DFe; 0.2  $\mu\text{m}$  filtered), and soluble iron (SFe; 0.02  $\mu\text{m}$  filtered) were investigated during the BONUS-GoodHope cruise in the Atlantic sector of the Southern Ocean (34°S/17°E–57°S/0°, February–March 2008). In the mixed layer, mean values of  $0.43 \pm 0.28$  and  $0.22 \pm 0.18$   $\text{nmol L}^{-1}$  were measured for TDFe and DFe, respectively. In deeper waters, TDFe and DFe concentrations were  $1.07 \pm 0.68$  and  $0.52 \pm 0.30$   $\text{nmol L}^{-1}$ , respectively. DFe concentrations decreased from the north (subtropical waters) to the south (Weddell Sea Gyre). In the subtropical domain, dusts coming from Patagonia and southern Africa and inputs from the African continental margin may explain high DFe and TDFe concentrations in surface and intermediate waters. Results from numerical models gave support to these hypotheses. In the Antarctic Circumpolar Current domain, estimation of the median advective time of water masses suggests that sediment inputs from the Antarctic Peninsula, South America margin, and/or South Georgia Islands could be an important source of Fe. Except in the subtropical domain where 0.4–0.6  $\text{nmol L}^{-1}$  of SFe were observed in the upper 1500 m, all stations exhibited values close to 0.1–0.2  $\text{nmol L}^{-1}$  in surface and 0.3–0.5  $\text{nmol L}^{-1}$  in deeper waters. For all stations, colloidal Fe (CFe) was a minor fraction of DFe in surface waters and increased with depth. Colloidal aggregation, sinking of CFe, and assimilation of SFe, followed by rapid exchange between the two fractions, are suspected to occur.

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## 1. Introduction

[2] The Southern Ocean is the largest high-nutrient low-chlorophyll (HNLC) area of the world ocean, characterized by low primary production along with excess nitrate. Since pioneering work by *Martin et al.* [1990], the pivotal role of iron (Fe) in such marine ecosystems has been demonstrated. This element, exhibiting very low concentrations in surface waters of the HNLC regimes ( $<0.1$   $\text{nmol L}^{-1}$ ), is an essential nutrient for all marine organisms, playing an important role in many biochemical reactions such as photosynthesis and nitrate reduction [e.g., *Sunda*, 1988–1989]. In situ and natural Fe fertilizations in the Southern Ocean have demonstrated

that Fe inputs enhance phytoplankton biomass and affect the major biogeochemical cycles (e.g., carbon (C) and nitrogen (N)) [*Coale et al.*, 2004; *Boyd et al.*, 2000, 2007; *Blain et al.*, 2007; *Pollard et al.*, 2009].

[3] The importance of new sources of Fe to the water column as well as the fractions that are truly bioavailable to the phytoplankton is still a question of debate. Atmospheric deposition was commonly thought to be the predominant external source of Fe in remote areas [*Jickells et al.*, 2005]. However, some studies have shown that inputs from sediments followed by upwelling or advection could provide important supply of Fe in surface waters of the open ocean [*Bucciarelli et al.*, 2001; *Elrod et al.*, 2004; *Lam and Bishop*, 2008; *Tagliabue et al.*, 2009a]. In the deep ocean, hydrothermal inputs could locally increase Fe concentrations [*Boyle and Jenkins*, 2008; *Bennett et al.*, 2008; *Tagliabue et al.*, 2010]. Finally, although considered as a minor source, iceberg melting could also supply Fe to surface waters [*Löscher et al.*, 1997; *Sedwick and DiTullio*, 1997; *Smith et al.*, 2007; *Lancelot et al.*, 2009].

[4] Most of the Fe supplied is present in the particulate pool ( $>0.2$   $\mu\text{m}$  [*de Baar and de Jong*, 2001]). Acidified unfiltered samples (total dissolvable iron (TDFe)) permit the study of the “labile” particulate pool that dissolves during acid storage

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[Löscher *et al.*, 1997] and gives information on the sources and distribution of iron in seawater [Chever *et al.*, 2010]. More than 99% of dissolved iron (DFe), which passes through a 0.2 or 0.4  $\mu\text{m}$  filter, is complexed by organic ligands [Gledhill and van den Berg, 1994; Rue and Bruland, 1995], preventing Fe from precipitation and removal by scavenging. The dissolved pool comprises colloidal Fe (CFe; 0.02–0.2  $\mu\text{m}$ ) and soluble Fe (SFe; <0.02  $\mu\text{m}$ ). The distribution of iron between these two pools (colloidal and soluble) appears related to the distribution of organic ligands that also exist as colloidal and soluble [Wu *et al.*, 2001]. Only a few studies have examined the soluble fraction of Fe, which exhibits nutrient-type profiles, suggesting that this is the bioavailable fraction [Wu *et al.*, 2001; Bergquist *et al.*, 2007]. However, although some studies demonstrated that SFe is more available, CFe can also be a source of Fe to phytoplankton [Chen and Wang, 2001; Chen *et al.*, 2003]. The distribution of these two pools of iron needs to be better investigated to further understand the biogeochemical cycle of Fe and its impact on phytoplankton and the C cycle.

[5] In this work, the distributions of DFe, TDFe, and SFe are studied along the BONUS-GoodHope transect, in the Atlantic sector of the Southern Ocean. This transect, from 34°S to 57°S, crossed contrasting oceanographic provinces from the subtropical waters of the southeast Atlantic Ocean to the eastern part of the Weddell Sea Gyre. We will discuss (1) the potential sources of DFe and TDFe in the different domains and (2) the distributions of SFe and CFe in the water column.

## 2. Materials and Methods

### 2.1. Study Area

[6] The BONUS-GoodHope cruise took place onboard the R/V Marion Dufresne in the Atlantic sector of the Southern Ocean from 8 February to 24 March 2008. Stations were sampled from the shelf region near Cape Town to 57°33'S along the Greenwich meridian. Figure 1 shows the main oceanographic fronts and domains crossed during the cruise, from north to south: (1) the subtropical domain and the southern subtropical front (S-STF), (2) the Antarctic Circumpolar Current (ACC) domain and its three major fronts (the sub-Antarctic front (SAF), the polar front (PF), and the southern ACC front (SACCF)), and (3) the eastern part of the Weddell Sea Gyre with the southern boundary (SBdy) separating this domain from the ACC.

[7] Twelve stations were sampled for total dissolvable iron (unfiltered iron), dissolved iron (<0.2  $\mu\text{m}$ ), and soluble iron (<0.02  $\mu\text{m}$ ). Among these stations, seven were sampled between 0 and 2000 m (large stations L1–L7), and five were sampled between 0 and 4000 m (super stations S1–S5). The position of each station is reported in Figure 1 and Table 1 along with the depth of the mixed layer (ML).

### 2.2. Sampling and Analyses

[8] Samples were collected with acid-cleaned 12 L Go-Flo bottles mounted on a Kevlar cable and tripped by a Teflon messenger, using trace metal clean protocols [Bruland *et al.*, 1979]. Sampling was performed in a clean container. All samples were collected in 60 ml acid-washed low-density polyethylene bottles. TDFe samples were collected without any filtration. Go-Flo bottles were then pressurized with

high-purity nitrogen allowing online filtration of seawater through 0.2  $\mu\text{m}$  filter cartridges (SARTOBRAN® 300, Sartorius) for DFe sampling. Soluble samples were obtained by immediate ultrafiltration of DFe on 0.02  $\mu\text{m}$  filters (ANOTOP 25®), which were previously acid cleaned with 10% Suprapur® hydrochloric acid (HCl; Merck) for 10 min and rinsed with milli-Q water for 5 min. This cleaning protocol avoids contamination of the SFe samples. However, we did not test other filters/membranes, and the soluble fraction that we present in this study is thus an operationally defined fraction, using the ANOTOP 25® aluminum oxide membrane. This needs to be taken into consideration when comparing our results to studies using other ultrafiltration methods.

[9] All samples were acidified with Ultrapur® HCl (Merck; 0.2% in volume for TDFe, final pH 1.7; 0.1% in volume for DFe and SFe, final pH 2.2). Samples were stored at room temperature and analyzed at the shore-based laboratory (LEMAR, Brest, France). Given that DFe samples were analyzed at the laboratory more than 2 months after sampling, we assume that the dissolved fraction represents the soluble fraction (SFe) plus the colloids that dissolved during acid storage [Chever *et al.*, 2010]. TDFe samples were stored for at least 5 months before analyses to release all but the most refractory particulate Fe species into the dissolved form [Löscher *et al.*, 1997].

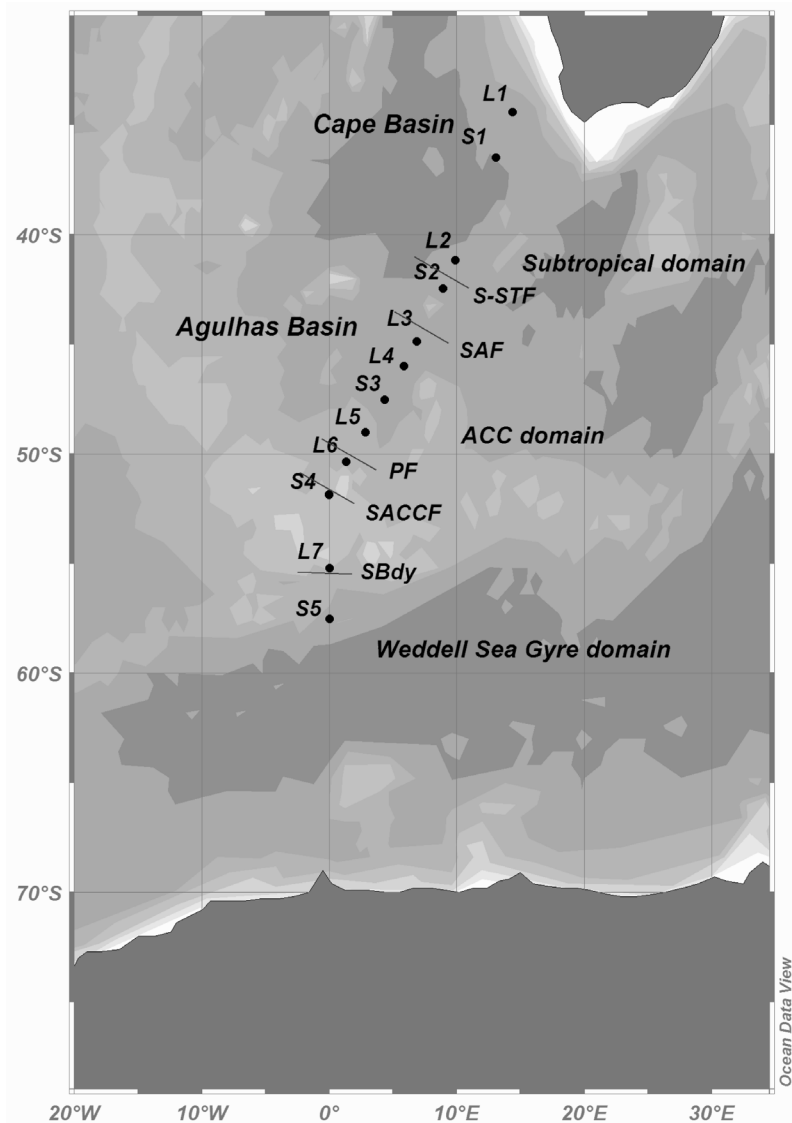
[10] Suprapur® ammonium (NH<sub>4</sub>OH; Merck) and ammonium acetate buffer purified three times through an 8-HQ resin were added to the samples to increase pH at 4.7, and analyses were performed by flow injection analysis (FIA) with online preconcentration onto 8-HQ resin and chemiluminescence detection [Obata *et al.*, 1993; modified by Sarthou *et al.*, 2003]. The detection limit was  $0.015 \pm 0.010 \text{ nmol L}^{-1}$  ( $n = 49$ ). The individual contributions to the total blank of Ultrapur® HCl (Merck), Suprapur® NH<sub>4</sub>OH (Merck), and ammonium acetate buffer were determined by addition of increasing amounts of these reagents to the sample and were lower than our detection limit. The accuracy of the method was assessed by analyzing the DFe reference materials collected during the SAFe intercomparison cruise (2004). Concentrations were equal to  $0.094 \pm 0.005 \text{ nmol L}^{-1}$  ( $n = 3$ ) and  $0.862 \pm 0.017 \text{ nmol L}^{-1}$  ( $n = 3$ ) for surface S1 and deep D2, respectively, in agreement with the certified values of  $0.097 \pm 0.043$  and  $0.91 \pm 0.17 \text{ nmol L}^{-1}$  [Johnson *et al.*, 2007].

## 3. Results

### 3.1. Hydrography

[11] Gladyshev *et al.* [2008] provided a description of the water mass arrangement along a track identical to that of the BONUS-GoodHope cruise, using hydrographic measurements performed in 2004. We briefly summarize the essential characters of this hydrological structure here, referring to the salinity section sampled during BONUS-GoodHope (Figure 2) and to the vertical profiles of temperature and salinity at all large and super stations (Figure 3).

[12] The subtropical domain extended southward to the S-STF (about 42°S), this limit being located between L2 and S2. The entire subtropical region is impacted by the Agulhas Current that follows the eastern coast of South Africa and retroflects eastward, forming intense anticyclonic eddies,



**Figure 1.** Location of the stations sampled during the BONUS-GoodHope cruise along with the three main oceanographic provinces encountered. The three domains crossed were the subtropical domain (stations L1, S1, and L2), the ACC domain (stations S2, L3, L4, S3, L5, L6, S4, and L7), and the eastern part of the Weddell Sea Gyre (station S5). Five fronts were crossed: the S-STF, SAF, PF, SACCF, and SBdy.

called Agulhas rings [Lutjeharms and van Ballegooyen, 1988; van Ballegooyen et al., 1994]. Its central water layer (potential temperature higher than  $\sim 10^{\circ}\text{C}$ , salinity higher than  $\sim 34.5$ ) (Figures 2, 3a, and 3b) was mostly occupied by water of Indian Ocean origin [Boebel et al., 2003]. At intermediate depths, two varieties of Antarctic Intermediate Water (AAIW) are generally found in this region [Gordon et al., 1992]. One closer to Africa with a salinity maximum exceeding 34.3 comes from the Indian Ocean through the Agulhas Current. The other one with a salinity maximum lower than 34.3 is formed in the Atlantic sector. The limit separating the two varieties was found near  $37^{\circ}\text{S}$  (south of S1) during the BONUS-GoodHope cruise. Below AAIW, the Upper Circumpolar Deep Water (UCDW) characterized by a core of low oxygen at  $\sim 1200$  m depth (and not distinguishable in Figure 2) did not show such easily separated

varieties, but its northern part was also likely conveyed by the Agulhas Current. Deeper along the subtropical segment of the cruise track, diluted North Atlantic Deep Water (NADW) was characterized by a salinity maximum at 2000–3000 m. The highest salinity values close to the African continental slope reflect advection by a southeastward deep boundary current [Arhan et al., 2003]. The deep-reaching turbulence causes this water mass to spread beyond the Agulhas Ridge. We refer to this variety of NADW as to southeast NADW. Finally, Antarctic Bottom Water (AABW) originating from the Weddell Sea present deeper than  $\sim 3500$  m in the Cape Basin (Figures 2, 3a, and 3b) is generally thought to circulate cyclonically in this basin [Reid, 1989].

[13] Further south, the domain of the ACC extended from the S-STF to the SBdy ( $\sim 42^{\circ}\text{S}$  to  $\sim 55^{\circ}\text{S}$ , stations L3–L7). This region is impacted by strong currents of westward origin.

**Table 1.** Location of the Stations Sampled During the Cruise in Relation to the Domains and Fronts Crossed and Mixed Layer Depth Observed From the Vertical Profile of Temperature for Nearby CTD Station

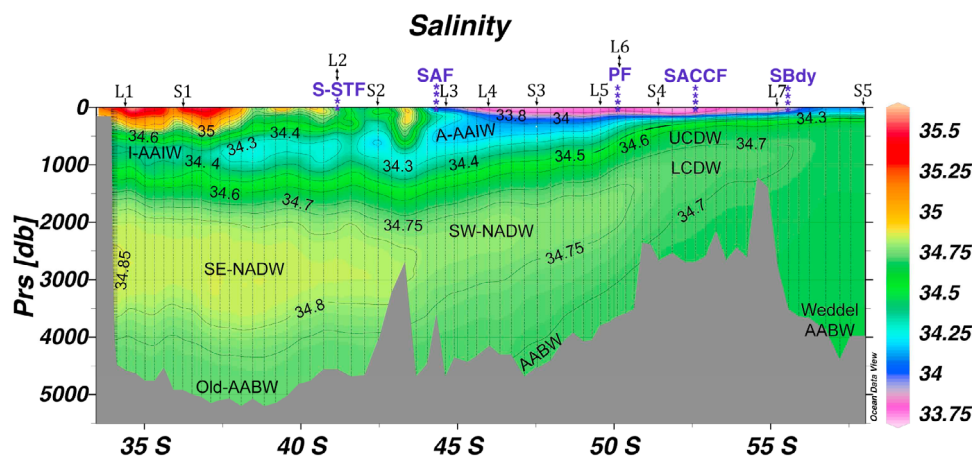
Oceanographic Domain	Fronts Crossed	Station	Position	ML Depth (m)
Subtropical		L1	34.43°S, 14.40°E	50–60
		S1	36.50°S, 13.10°E	40–50
		L2	41.18°S, 09.92°E	25
ACC	S-STF		42.2°S	
		S2	42.47°S, 08.93°E	50–80
	SAF		44.2°S	
		L3	44.88°S, 06.88°E	60–80
		L4	46.02°S, 05.87°E	80
	PF	S3	47.55°S, 04.37°E	80–100
		L5	49.03°S, 02.84°E	100–110
	SACCF		50.2°S	
		L6	50.38°S, 01.33°E	60–80
			51.5°S	
SBdy	S4	51.85°S, 00.00°E	120–150	
	L7	55.23°S, 00.03°E	80–110	
Eastern part of the Weddell Sea Gyre		S5	57.55°S, -00.03°E	100

Although station S2 is located south of the S-STF, its surface waters exhibit salinity and temperature signatures of subtropical waters, which suggest an advection from the S-STF region by a neighboring eddy. As S2 otherwise appears like ACC stations in terms of DFe distribution (Figure 4a), it will be discussed in this domain (see section 4.1.2). The SAF and PF were found at  $\sim 45^\circ\text{S}$  and  $51^\circ\text{S}$ , respectively. Between these two fronts, the water mass with salinity lower than 34.3 was AAIW. It was located beneath the surface mixed layer and winter water (marked by a temperature minimum in Figure 3c). Below AAIW were UCDW, the deep waters visible from their southward shallowing salinity maximum, and the Antarctic Bottom Waters here observed against the northern flank of the Mid-Atlantic Ridge (MAR). *Whitworth and Nowlin* [1987] showed that the deep waters at these latitudes are composed of Lower Circumpolar Deep Water with, north of the PF, an addition of diluted southwest NADW, which flows along the continental slope of South America (Argentinean Basin) before being injected in the ACC in the southwestern Atlantic.

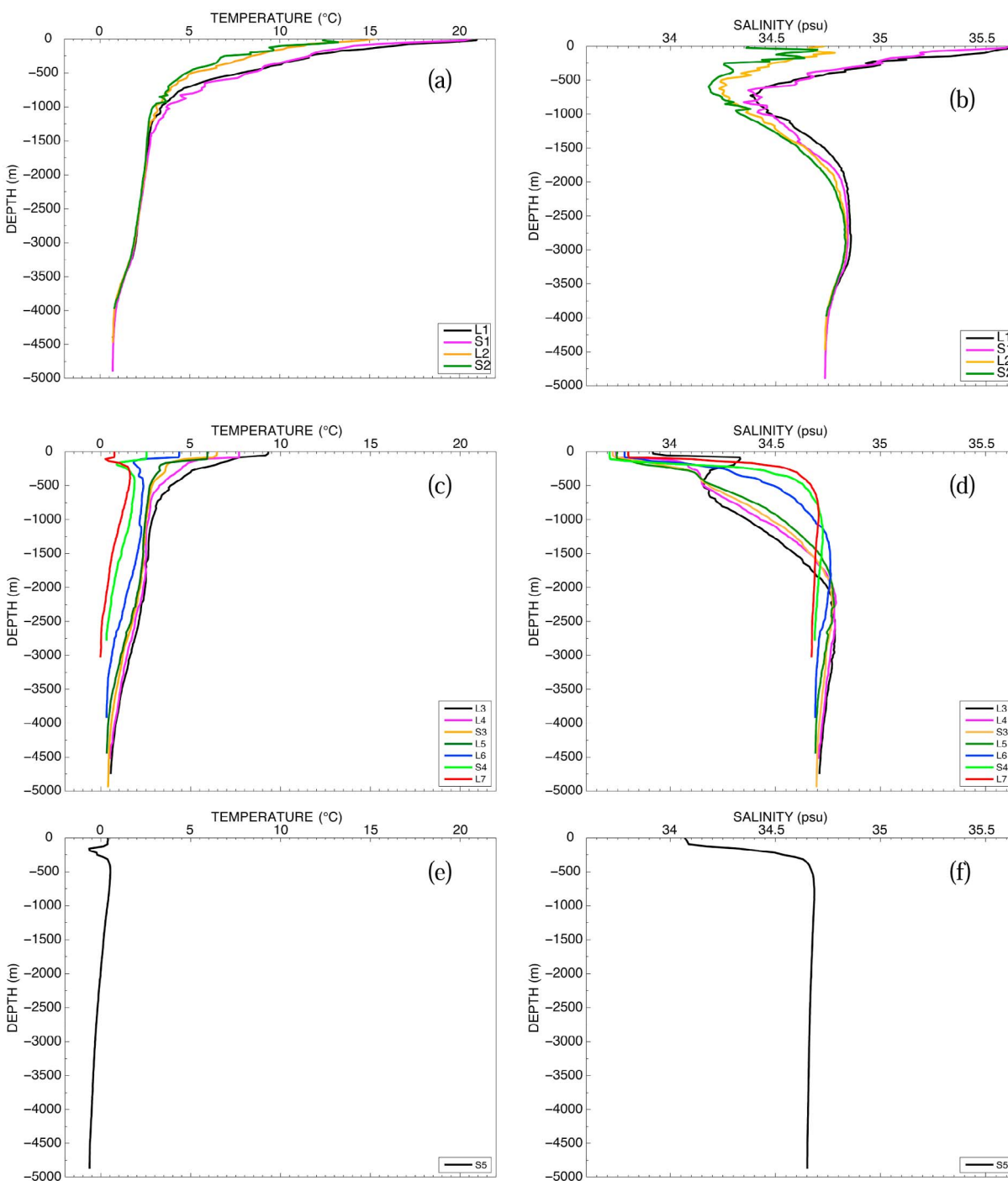
[14] South of the SBdy, and only sampled at S5, were waters entrained in the large-scale cyclonic flow of the Weddell Sea Gyre. The near-surface waters are thought to have been in contact with the western continental margin of the Antarctic Peninsula, while the deeper waters might have had a more recent contact with the northern topographic limit of the Weddell Basin [*Orsi et al.*, 1993; *Meredith et al.*, 2000; *Klatt et al.*, 2005].

### 3.2. Fe Distributions

[15] Concentrations of DFe, TDFe, and SFe are reported in Table 2. Apparent particulate iron ( $\text{Fe}_{\text{app}}$ ), calculated by subtracting DFe from TDFe, is defined as biogenic iron and labile lithogenic particulate Fe, and iron adsorbed on lithogenic or biogenic particles [*de Baar et al.*, 1999]. Mean values of DFe, TDFe, and  $\text{Fe}_{\text{app}}$  in the mixed layer and below the mixed layer are presented in Table 3. The sections of DFe and  $\text{Fe}_{\text{app}}$  are presented in Figure 4. These sections have, however, to be interpreted with caution given the extrapolation caused by the large distance between some stations.



**Figure 2.** Vertical distribution of salinity (psu) encountered along the transect from the north (left) to the south (right) of the section. Water masses are indicated. I-AAIW, Indian Antarctic Intermediate Water; A-AAIW, Atlantic Antarctic Intermediate Water; SE-NADW, southeast North Atlantic Deep Water; SW-NADW, southwest North Atlantic Deep Water; LCDW, Lower Circumpolar Deep Water.

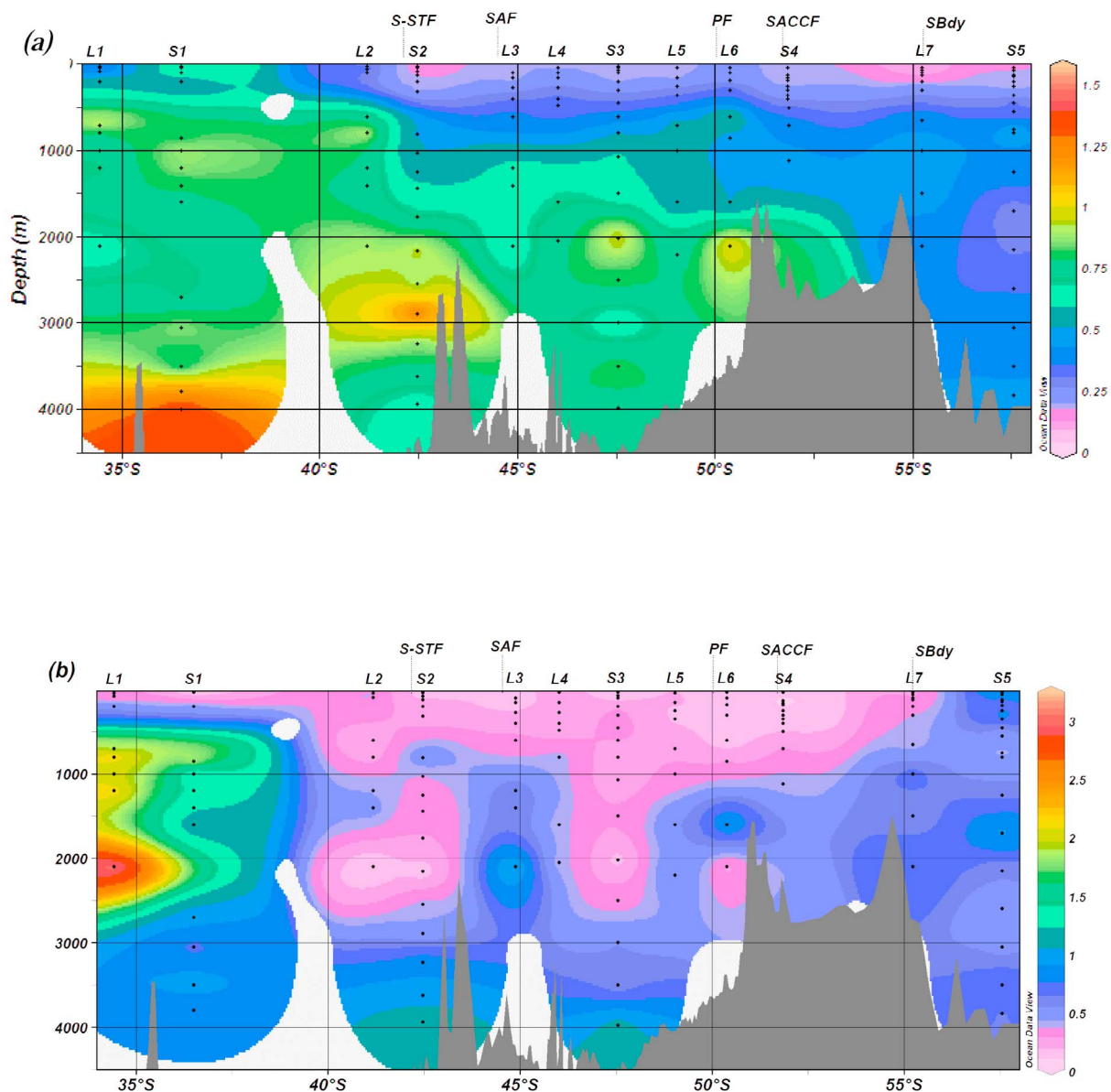


**Figure 3.** Vertical profiles of potential (a, c, and e) temperature ( $^{\circ}\text{C}$ ) and (b, d, and f) salinity (psu) in the three different domains. Stations located in the same domain are considered together.

[16] Figure 4a shows a decrease in DFe concentrations from the north to the south of the section. In the ML, values ranged from  $0.064 \pm 0.006 \text{ nmol L}^{-1}$  (L7, 100 m) to  $0.720 \pm 0.011 \text{ nmol L}^{-1}$  (S1, 30 m). Below the ML, values ranged from  $0.092 \pm 0.004 \text{ nmol L}^{-1}$  (L7, 120 m) to  $1.583 \pm 0.009 \text{ nmol L}^{-1}$  (S1, 4000 m).

[17] TDFe values were the lowest in the ACC domain. In the ML, values ranged from  $0.191 \pm 0.007 \text{ nmol L}^{-1}$  (S4, 130 m) to  $1.276 \pm 0.027 \text{ nmol L}^{-1}$  (S5, 60 m). Below the ML,

concentrations ranged from  $0.234 \pm 0.012 \text{ nmol L}^{-1}$  (S4, 160 m) to  $3.918 \pm 0.063 \text{ nmol L}^{-1}$  (L1, 2100 m). The section of  $\text{Fe}_{\text{app}}$  is presented in Figure 4b. In the ML, concentrations were low, ranging from  $0.025 \pm 0.027 \text{ nmol L}^{-1}$  (S3, 70 m) to  $0.424 \pm 0.058 \text{ nmol L}^{-1}$  (L1, 20 m), except at the most southerly station (S5) where values were close to  $1 \text{ nmol L}^{-1}$ . Below the ML, the highest  $\text{Fe}_{\text{app}}$  concentrations were measured in the subtropical domain between 800 and 2100 m, with values ranging from  $1.944 \pm 0.110$  to  $3.258 \pm$



**Figure 4.** Vertical section of (a) DFe and (b)  $\text{Fe}_{\text{app}}$  concentrations (in  $\text{nmol L}^{-1}$ ) from the north (left) to the south (right) of the section. Note that the concentration scale is not the same.

$0.071 \text{ nmol L}^{-1}$ . In the ACC and at S5 station,  $\text{Fe}_{\text{app}}$  values varied from  $0.026 \pm 0.060 \text{ nmol L}^{-1}$  (L6, 600 m) to  $1.252 \pm 0.047 \text{ nmol L}^{-1}$  (L3, 2100 m). In the ML,  $\text{Fe}_{\text{app}}$  represented  $42 \pm 23\%$  ( $n = 20$ ) of the TDFe except at S5, where it represented  $88 \pm 1\%$  ( $n = 29$ ) of the TDFe. Below the ML,  $\text{Fe}_{\text{app}}$  represented  $80 \pm 4\%$  ( $n = 4$ ) of the TDFe at the L1 station between 800 and 2100 m, whereas it represented  $47 \pm 18\%$  ( $n = 109$ ) in the remaining transect.

[18] Vertical profiles of DFe, SFe, and colloidal Fe ( $\text{CFe} = \text{DFe} - \text{SFe}$ ) are plotted in Figures 5a–5c for the five super stations. Except at the S1 station where high values of SFe were observed in the upper 1500 m ( $0.4\text{--}0.6 \text{ nmol L}^{-1}$ ) (Figure 5b), all stations exhibited the same pattern with values close to  $0.1\text{--}0.2 \text{ nmol L}^{-1}$  in surface and  $0.3\text{--}0.5 \text{ nmol L}^{-1}$  in deeper waters. For all stations, CFe was the minor fraction of DFe in surface (i.e.,  $23 \pm 11\%$ ,  $n = 3$ ), but this fraction increased with depth (i.e.,  $37 \pm 21\%$  of the DFe ( $n = 28$ ))

between the ML and 2000 m and  $53 \pm 15\%$  ( $n = 16$ ) below 2000 m).

#### 4. Discussion

[19] DFe concentrations observed during BONUS-GoodHope are in the same range as previous values measured in the same sector of the Southern Ocean by *Landing and Powell* [1998] ( $33^\circ\text{S}$ ,  $0.5\text{--}2.6 \text{ nmol L}^{-1}$ ) and *de Jong et al.* [1999] ( $\sim 52^\circ\text{--}60^\circ\text{S}$ ,  $0.05\text{--}0.90 \text{ nmol L}^{-1}$ ). The decrease in concentrations from the north to the south is consistent with a recent study south of Tasmania (SR3 section, between  $42^\circ\text{S}$  and  $54^\circ\text{S}$ ) during fall 1998 [*Sedwick et al.*, 2008]. TDFe concentrations are coherent with values measured in the same sector of the Southern Ocean ( $6^\circ\text{E}$ ,  $50^\circ\text{--}60^\circ\text{S}$ ,  $0.18\text{--}2.69 \text{ nmol L}^{-1}$ ) [*Croot et al.*, 2004].

**Table 2.** DFe, TDFe, and SFe and Location of the Stations<sup>a</sup>

Domain	Station	Position	Bottom Depth (m)	Depth (m)	DFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )	TDFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )	SFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )			
Subtropical	L1	34.43°S, 14.40°E	4505	20	0.230	0.008	0.654	0.050					
				40	0.501	0.002	0.678	0.014					
				60	nv		0.547	0.022					
				80	0.211	0.002	0.400	0.012					
				200	0.745	0.002	1.469	0.067					
				700	1.155	0.012	2.740	0.156					
				800	0.639	0.006	3.320	0.109					
				1000	0.656	0.008	2.600	0.102					
				1200	0.633	0.006	3.202	0.174					
				2100	0.660	0.008	3.918	0.063					
				S1	36.50°S, 13.10°E	4915	20	0.700	0.007	0.730	0.012	nv	
							30	0.720	0.011	0.767	0.032	0.525	0.006
	40	nv					0.817	0.015	0.425	0.022			
	70	nv					0.575	0.016					
	100	0.815	0.022				nv		nv				
	200	0.364	0.005				0.968	0.040					
	300	nv					2.248	0.074	0.563	0.049			
	500	nd					1.971	0.013					
	700	nv					2.727	0.037	0.646	0.010			
	850	0.717	0.014				3.185	0.049					
	1000	1.014	0.037				1.369	0.036					
	1200	0.943	0.005				2.380	0.088	0.628	0.007			
	1400	0.764	0.010				1.881	0.011					
	1600	0.724	0.112				1.619	0.022	0.366	0.015			
	2000	nd					1.728	0.033					
	2700	0.658	0.012				1.621	0.022					
	3050	0.911	0.002				1.536	0.020	0.368	0.003			
	3500	0.585	0.005				1.683	0.010					
	3800	1.154	0.011				1.838	0.025	0.448	0.006			
	4000	1.583	0.009	nd									
	L2	41.18°S, 09.92°E	4525	15	0.156	0.000	0.293	0.013					
				35	nv		0.349	0.008					
				45	0.590	0.054	1.261	0.012	0.431	0.082			
95				0.291	0.013	0.729	0.021						
300				nv		0.455	0.011						
600				0.741	0.010	0.934	0.018						
800				1.098	0.032	1.265	0.039	0.266	0.004				
1200				0.829	0.008	1.277	0.026						
1400				0.776	0.024	1.350	0.009						
2100				0.915	0.040	0.987	0.404						
ACC				S2	42.47°S, 08.93°E	4070	15	0.179	0.013	0.265	0.016	nv	
							30	0.129	0.012	0.284	0.010	nv	
	35	0.098	0.008				0.196	0.008	nv				
	45	nd					nd						
	80	0.152	0.004				0.292	0.004	nv				
	120	0.164	0.005				0.314	0.010	0.162	0.012			
	196	0.178	0.005				0.624	0.018	nv				
	314	0.169	0.004				0.412	0.004					
	461	nv					0.604	0.019	0.200	0.009			
	598	nv					0.849	0.037					
	809	0.434	0.003				1.123	0.014					
	1029	0.431	0.007				0.736	0.001	0.319	0.004			
	1250	0.460	0.006				0.823	0.020	0.364	0.008			
	1441	0.641	0.010				0.834	0.016	0.415	0.009			
	1764	0.649	0.005				0.950	0.020					
	2156	1.028	0.014				1.069	0.034	0.369	0.005			
	2548	0.785	0.014				1.240	0.025	0.378	0.007			
	2891	1.459	0.015				1.828	0.048	0.330	0.003			
	3234	0.782	0.026				1.714	0.022					
	3626	0.716	0.010				1.687	0.064	0.339	0.006			
	3940	0.625	0.008				1.840	0.033					
L3	44.88°S, 06.88°E	4315	30	nv		0.265	0.019	0.215	0.008				
			100	0.205	0.018	0.279	0.010	nv					
			150	0.121	0.011	0.268	0.017	0.097	0.021				
			270	0.203	0.005	0.376	0.011						
			400	0.337	0.013	0.651	0.007	0.327	0.021				
			600	0.435	0.007	0.759	0.011						
			1200	0.613	0.022	1.248	0.037						
			1400	0.621	0.008	1.283	0.014	0.335	0.012				
			2100	0.600	0.002	1.852	0.046	0.339	0.014				



Table 2. (continued)

Domain	Station	Position	Bottom Depth (m)	Depth (m)	DFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )	TDFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )	SFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )
	L4	46.02°S, 05.87°E	4147	30	0.165	0.017	0.477	0.069		
				60	nv		0.276	0.003	0.103	0.010
				100	0.208	0.011	0.345	0.022		
				150	0.229	0.013	0.441	0.005		
				270	0.280	0.006	0.582	0.012	0.119	0.001
				400	0.346	0.006	0.487	0.026		
				480	0.381	0.026	0.637	0.010	0.145	0.012
				800	0.331	0.004	0.806	0.028	0.239	0.002
				1600	0.699	0.027	1.034	0.026	0.150	0.002
				2050	0.782	0.019	1.090	0.020	0.236	0.007
	S3	47.55°S, 04.37°E	4480	20	0.161	0.020	0.318	0.016	0.144	0.005
				30	0.170	0.004	0.245	0.015		
				40	nv		0.261	0.013	0.186	0.004
				70	0.185	0.014	0.210	0.013		
				100	0.187	0.021	0.324	0.010		
				200	0.141	0.009	0.629	0.011	0.140	0.004
				300	0.281	0.003	0.636	0.003		
				450	0.286	0.000	0.651	0.048	nv	
				600	0.410	0.011	0.706	0.008		
				800	0.557	0.011	0.735	0.009	0.283	0.029
				1070	0.656	0.014	0.843	0.018	0.268	0.020
				1500	0.600	0.005	0.882	0.012	0.284	0.007
				2020	1.096	0.034	1.156	0.074		
				2500	0.796	0.030	1.102	0.166		
				3000	0.591		1.168	0.077	0.263	0.020
				3500	0.825	0.006	1.345	0.049	0.300	0.002
				3980	0.761	0.017	1.946	0.096	0.340	0.002
	L5	49.03°S, 02.84°E	4025	40	0.141	0.005	0.360	0.034		
				80	nv		0.381	0.284		
				150	0.133	0.001	0.383	0.014		
				170	nv		0.238	0.006	nv	
				250	0.216	0.001	0.356	0.001	nv	
				350	0.354	0.015	0.519	0.018		
				700	0.604	0.012	0.905	0.006		
				1000	0.504	0.003	0.812	0.006	0.425	0.151
				1600	0.473	0.007	1.051	0.027	nv	
				2200	0.722	0.016	1.323	0.014	0.517	0.015
	L6	50.38°S, 01.33°E	3576	30	0.249	0.016	0.275	0.020	nv	
				60	nv		0.300	0.034		
				100	0.217	0.023	0.455	0.016	nv	
				135	nv		0.362	0.013	nv	
				180	0.211	0.009	0.402	0.009		
				300	0.421	0.074	0.598	0.009	0.304	0.020
				600	0.850	0.023	0.876	0.037		
				850	0.449	0.009	0.775	0.013	0.380	0.002
				1600	0.431	0.013	1.470	0.043		
				2100	1.123	0.035	1.260	0.054	0.566	0.017
	S4	51.85°S, 00.00°E	2632	30	0.182	0.004	0.338	0.028	0.128	0.018
				60	nv		0.240	0.011		
				130	0.136	0.008	0.191	0.007	nv	
				160	0.126	0.006	0.234	0.012	nv	
				180	0.184	0.002	0.264	0.007		
				250	0.191	0.005	0.258	0.005	nv	
				300	0.202	0.012	0.309	0.002		
				350	0.203	0.003	0.373	0.008		
				400	0.213	0.005	0.360	0.007	nv	
				500	0.447	0.002	0.708	0.005	0.334	0.015
				700	0.426	0.002	0.792	0.047		
				900	nv		0.546	0.042	0.287	0.008
				1117	0.460	0.003	0.885	0.034		
				1950	nv		0.799	0.022	0.290	0.016
				2300	nv		1.510	0.022	0.335	0.006
				2500	0.787	0.027	1.240	0.021	0.339	0.024
	L7	55.23°S, 00.03°E	2770	30	0.110	0.002	0.381	0.017		
				60	0.065	0.004	0.224	0.021	nv	
				100	0.064	0.006	0.244	0.007		
				120	0.092	0.004	0.549	0.014	nv	
				200	0.212	0.003	0.524	0.007		
				300	0.369	0.003	0.794	0.084		
				650	0.451	0.002	0.810	0.010	nv	

**Table 2.** (continued)

Domain	Station	Position	Bottom Depth (m)	Depth (m)	DFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )	TDFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )	SFe (nmol L <sup>-1</sup> )	STD (nmol L <sup>-1</sup> )
Eastern part of the Weddell Sea Gyre	S5	57.55°S, -00.03°E	3932	1000	0.494	0.007	1.239	0.007		
				1500	0.482	0.010	1.044	0.017	0.282	0.014
				2100	0.390	0.009	1.171	0.054	0.326	0.017
				30	0.145	0.011	1.231	0.031	nv	
				60	0.144	0.001	1.276	0.027		
				120	0.313	0.008	0.894	0.037	0.245	0.021
				140	0.110	0.010	0.637	0.027		
				190	0.133	0.002	0.779	0.006	nv	
				250	0.166	0.005	1.057	0.005	nv	
				350	0.142	0.001	1.303	0.019		
				450	0.517	0.029	0.758	0.014	0.326	0.017
				550	0.161	0.004	0.657	0.024		
				750	0.713	0.040	0.933	0.019	0.268	0.012
				800	0.341	0.005	0.895	0.018		
				1250	0.417	0.007	1.030	0.017	0.214	0.008
				1700	0.259	0.004	1.308	0.081		
				2150	0.290	0.003	0.803	0.013		
2600	0.365	0.016	0.818	0.108						
3050	0.418	0.003	0.909	0.033	0.237	0.001				
3500	0.382	0.003	1.174	0.017						
3840	0.511	0.023	1.121	0.015	0.212	0.016				

<sup>a</sup>Uncertainties on the concentrations correspond to standard deviation (SD) of a same sample measured three times. SFe samples were not validated when concentrations were higher than DFe. DFe samples were not validated when concentrations were higher than TDFe. Here nv, not validated; nd, not determined.

[20] Vertical profiles of SFe were rarely studied in the Southern Ocean and were analyzed in geographically restricted areas in the Atlantic Ocean. To our knowledge, only one profile was reported in the Southern Ocean during the EisenEx iron enrichment experiment, outside the iron-fertilized patch (~20°E, 48°S) [Nishioka *et al.*, 2005]. Concentrations were twice lower than those measured in our study, with mean concentrations of  $0.04 \pm 0.01$  nmol L<sup>-1</sup> ( $n = 5$ ) and  $0.16 \pm 0.03$  nmol L<sup>-1</sup> ( $n = 9$ ) for surface and deep waters, respectively. In the Pacific and Atlantic Oceans, SFe concentrations were equal to ~0.1 nmol L<sup>-1</sup> in the surface and to ~0.3–0.4 nmol L<sup>-1</sup> in deeper water [Wu *et al.*, 2001; Bergquist *et al.*, 2007].

[21] In the surface layer of the North Pacific and Atlantic Oceans, contrary to what we observed, CFe represented the main fraction of DFe (71–90%) [Nishioka *et al.*, 2001; Wu *et al.*, 2001; Cullen *et al.*, 2006]. However, the percentage of CFe was much lower (~35%) during the EisenEx enrichment in the Southern Ocean [Nishioka *et al.*, 2005]. In the deep water, the proportion of CFe relative to DFe observed during our study was similar to values observed in the literature (30–70%) [Wu *et al.*, 2001; Cullen *et al.*, 2006; Nishioka *et al.*, 2005].

#### 4.1. Sources of Iron

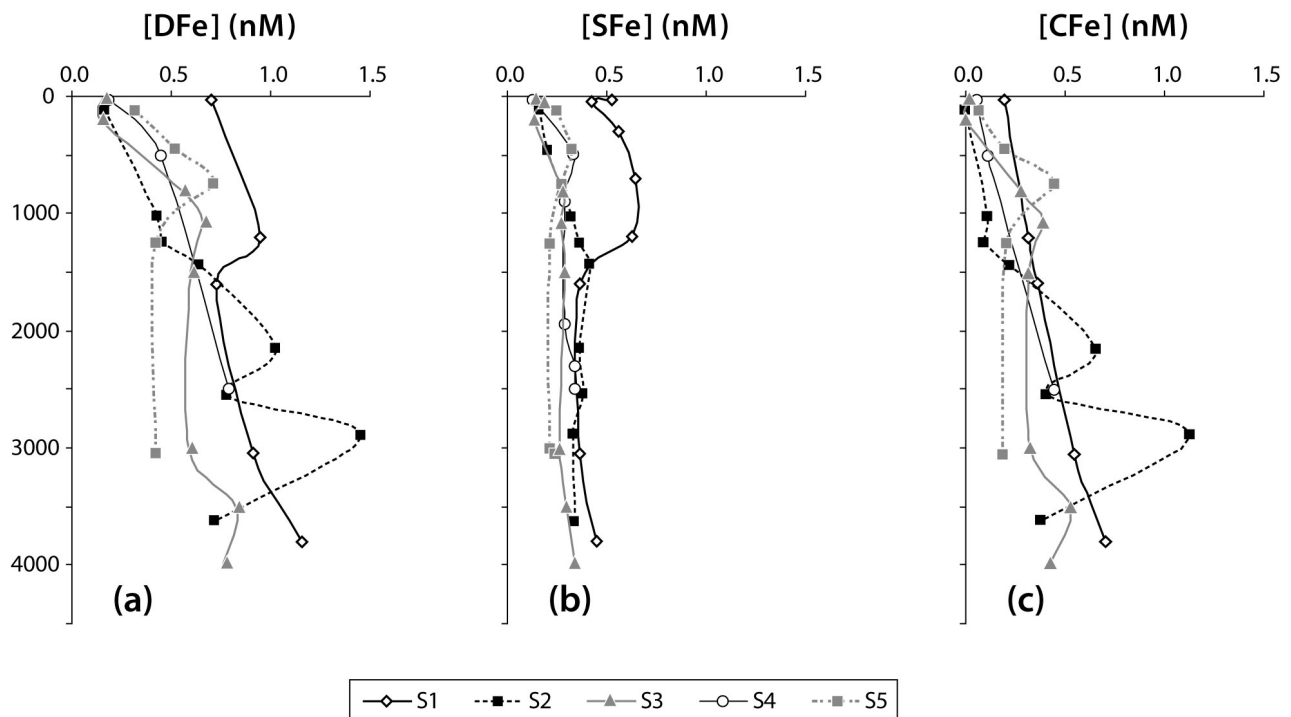
##### 4.1.1. Subtropical Domain (Stations L1, S1, and L2)

[22] The atmospheric input is a potential source of Fe in this region. This input is recognized to be temporally and spatially variable with some sporadic strong inputs [Jickells and Spokes, 2001]. Several studies suggest that the Patagonian desert could be one of the main sources of dust in the Atlantic sector of the Southern Ocean [Mahowald *et al.*, 1999; Erickson *et al.*, 2003; Meskhidze *et al.*, 2007], with higher dust activity generally observed in the summer period [Gassó and Stein, 2007]. The NOAA Hybrid Single-Particle

Lagrangian Integrated Trajectory (HYSPPLIT) model (<http://www.arl.noaa.gov/ready.html>) allows us to determine the potential origin of air masses during the cruise at a determined point. This air mass back trajectory model was run for all of the stations at heights of 10, 500, and 1000 m over a 5 day period. Air masses originating from Patagonia were observed during the cruise only at the three subtropical stations (Figure 6). However, this model gives us information on a short time scale (few days). To assess the cumulative aeolian input of DFe over a seasonal time scale (4 months) in the subtropical domain, we used a regional oceanic model based on the Regional Ocean Modelling System (ROMS) (Southern African Experiments (SAFe) [Penven *et al.*, 2006]) forced by two different atmospheric dust products. ROMS-SAFE resolves the complex physics of the region, but this model does not include any DFe loss terms (e.g., scavenging/precipitation or biotic uptake) and should therefore be seen to provide the maximum potential impact of dust Fe on DFe. To account for DFe losses, we also used results of the global ocean biogeochemistry model Pelagic Interaction Scheme for Carbon and Ecosystem Studies (PISCES) when dust DFe sources were eliminated during equilibrium simulations. PISCES is a lower resolution than ROMS-SAFE but provides an estimate as to the overall importance of different DFe

**Table 3.** Mean Concentrations of DFe, TDFe, and Fe<sub>app</sub> in the ML and Below the ML

	Position	Mean Value
DFe (nmol L <sup>-1</sup> )	ML	$0.216 \pm 0.177$ ( $n = 23$ )
	Below ML	$0.521 \pm 0.303$ ( $n = 113$ )
TDFe (nmol L <sup>-1</sup> )	ML	$0.430 \pm 0.283$ ( $n = 31$ )
	Below ML	$1.066 \pm 0.684$ ( $n = 127$ )
Fe <sub>app</sub> (nmol L <sup>-1</sup> )	ML	$0.234 \pm 0.300$ ( $n = 22$ )
	Below ML	$0.559 \pm 0.551$ ( $n = 113$ )



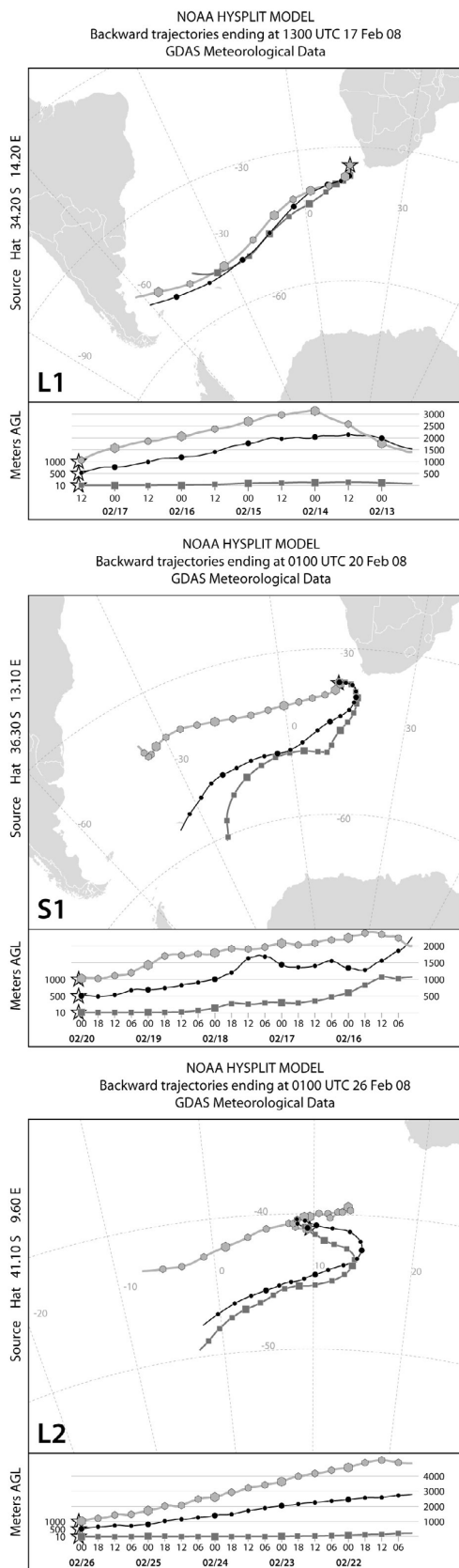
**Figure 5.** Vertical profiles of (a) DFe, (b) SFe, and (c) CFe (in  $\text{nmol L}^{-1}$ ) at the five super stations.

sources when the iron cycle is accounted for [Tagliabue *et al.*, 2009a]. We used two different estimates of atmospheric dust input to the ocean. The first is the National Center for Atmospheric Research (NCAR) model [Mahowald *et al.*, 2006], which has a large Patagonian dust source ( $\sim 20 \text{ mg m}^{-2} \text{ month}^{-1}$  close to Patagonia,  $2\text{--}10 \text{ mg m}^{-2} \text{ month}^{-1}$  around southern Africa, and  $2\text{--}5 \text{ mg m}^{-2} \text{ month}^{-1}$  along the cruise transect in the subtropical domain). The second is the Integrated Catchment Model (INCA) model [Aumont *et al.*, 2008], which has a reduced Patagonian source but a greater source from southern Africa ( $< 2 \text{ mg m}^{-2} \text{ month}^{-1}$  close to Patagonia,  $2\text{--}50 \text{ mg m}^{-2} \text{ month}^{-1}$  around southern Africa, and  $\sim 2 \text{ mg m}^{-2} \text{ month}^{-1}$  along the cruise transect in the subtropical domain). A Fe content of 3.5% [Desboeufs *et al.*, 2005] and a solubility of 5% [Baker *et al.*, 2006] and 10% [Duce and Tindale, 1991] were applied to each dust model for study with ROMS-SaFe (PISCES simulations used their typical experimental design [see Tagliabue *et al.*, 2009a]). The solubility of 10% used with ROMS-SaFe also agrees with the average value of 10.3% calculated for samples collected in the South Atlantic south of  $35^\circ\text{S}$  (A. Baker, personal communication, 2010).

[23] Using NCAR, ROMS-SaFe suggests that at the two groups of stations (L1/S1 and L2), dust deposition from Patagonia could be a significant source of DFe (up to  $\sim 0.6 \text{ nmol L}^{-1}$  after 4 months) (Figure 7b, dotted lines). Similarly, the results from PISCES suggest that up to  $0.2 \text{ nmol L}^{-1}$  of DFe at these stations was maintained by dust deposition from Patagonia at steady state. However, stations L1, S1, and L2 might also have been enriched by atmospheric dust inputs from southern Africa, followed by lateral advection. Indeed, major transport pathways of air masses over southern Africa are from the interior of the continent toward the Indian Ocean [Piketh *et al.*, 2002]. Experiments with

ROMS-SaFe using INCA dust deposition showed that at L1 and S1 stations, the maximum potential enrichment was around  $0.15$  or  $0.3 \text{ nmol L}^{-1}$  after 4 months, using 5% or 10% of solubility, respectively (Figures 7a and 7b, solid line). Analyzed more in detail, the model results revealed that the increase of  $0.15\text{--}0.3 \text{ nmol L}^{-1}$  was mainly controlled by the advection of Indian Ocean waters (data not shown). At the L2 station, there was no significant advection of Indian Ocean waters (increase in DFe  $< 0.05 \text{ nmol L}^{-1}$  after 4 months) (Figures 7a and 7b, dotted line), suggesting that only direct dust deposition coming from Patagonia is likely to increase DFe in surface waters at this station. In accord, under INCA dust forcing, PISCES suggests that dust contributes  $0.1 \text{ nmol L}^{-1}$  to DFe at L1 and S1, with a contribution of  $< 0.05 \text{ nmol L}^{-1}$  at L2.

[24] Another potential source of Fe to the subtropical region is the continental margin. Indeed, the surface waters of the Cape Basin from Indian Ocean origin might also have been enriched by Fe when they swept the African continental margin. Previous work in the Southern Ocean already mentioned regions impacted by advection of water masses that have been in contact with continental shelves [Sedwick *et al.*, 1997]. The transit time of the water masses between the southeast Indian Ocean ( $30^\circ\text{S}$ ) and the BONUS-GoodHope transect was estimated from the Archiving, Validation, and Interpretation of Satellite Oceanographic Mean Absolute Dynamic Topography [Ducet *et al.*, 2000] for both the specific cruise period (2007–2008) and for the entire satellite time series (i.e., from 1992 to 2008). It gave an estimate for the mean advective time between 1 and 3 months. Given that the residence time of Fe in surface water is on the order of days to months [de Baar and de Jong, 2001; Croot *et al.*, 2004; Sarthou *et al.*, 2003], Fe transported from the Indian Ocean could be seen in this domain. PISCES results



**Figure 6.** The 5 day back trajectory analysis for heights of 10, 500, and 1000 m, starting from L1, S1, and L2 stations.

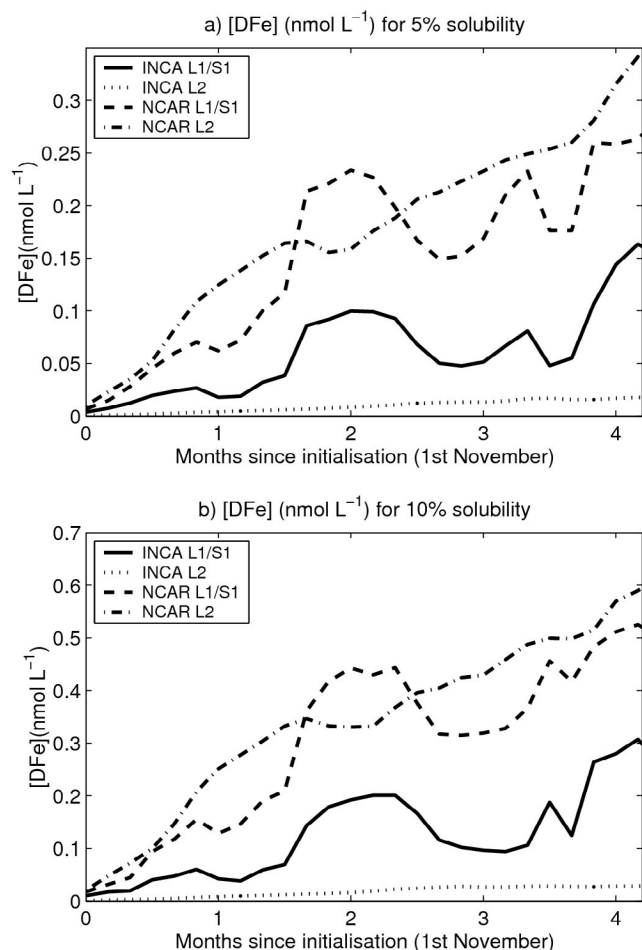
also suggest that this source can contribute between 0.1 and 0.3 nmol L<sup>-1</sup> at stations L2 and L1, respectively (see *Tagliabue et al.* [2009a] for experimental design). As such, these impacts are of similar order to those arising from atmospheric inputs.

[25] Below the surface waters, the AAIW of Indian Ocean origin was observed as deep as 850 m (Figure 2) at the L1 and S1 stations. Those water masses could also be impacted by the African eastern and southern continental slopes, explaining higher DFe concentrations at these subtropical stations (0.6–0.7 nmol L<sup>-1</sup>) than southerly (<0.5 nmol L<sup>-1</sup> in the ACC domain). Particles largely dominated the Fe pool in the subtropical domain and particularly at the L1 station in the UCDW and southward slope NADW current. High particle concentrations in the vicinity of the margins have already been observed [*Löscher et al.*, 1997; *Elrod et al.*, 2004; *Lam and Bishop*, 2008]. The decrease in Fe<sub>app</sub> concentrations from L1 to L2 also agrees with this continental source.

[26] At the S1 station, in the AABW, high DFe values (1.15 and 1.58 nmol L<sup>-1</sup>) were observed. The water mass at the bottom of the Cape Basin indeed originates from the Weddell Sea and likely transited through fractures in the MAR. Such a mechanism of Fe enrichment at the bottom of the water column was already observed by *Sedwick et al.* [1997] and *Laës et al.* [2003] in the Australian sub-Antarctic region and in the eastern North Atlantic waters, respectively.

#### 4.1.2. ACC Domain (Stations S2, L3, L4, S3, L5, L6, S4, and L7)

[27] The decrease in DFe concentrations from the subtropical domain to the ACC could be due to lower Fe inputs. In the ACC region, the NOAA HYSPLIT model did not suggest any air mass coming from Patagonia or southern Africa during the 5 day period before sampling. The main source of Fe may thus be advection of water masses from the west through the ACC, enriched by Fe from the sediments of the Antarctic Peninsula, South America margin, and/or South Georgia Islands [*Arhan et al.*, 2002; *Löscher et al.*, 1997; *Croot et al.*, 2004; *Korb et al.*, 2004]. Mean water velocities across the ACC have been estimated in the Drake Passage between 2004 and 2007 to be on the order of  $16.7 \pm 0.2 \text{ cm s}^{-1}$  [*Cunningham and Pavic*, 2007]. Estimates of the median advection time for these water masses using outputs of a global ocean general circulation model (ORCA05) and a Lagrangian quantitative diagnostic (ARIANE, <http://stockage.univ-brest.fr/~grima/Ariane/ariane.html>) [*Blanke et al.*, 2006]) suggest a mean transit time of 2 years with a minimum value of 1 year. The residence time of DFe is on the order of days to months in surface waters and of 15–40 years in deep waters [*de Baar and de Jong*, 2001; *Croot et al.*, 2004; *Sarthou et al.*, 2003]. The imprint of Fe coming from South America, the Antarctic Peninsula, and the South Georgia Islands is thus more likely to be seen in the deep waters than in the surface ones. Moreover, the lowest Fe<sub>app</sub> concentrations of the section were found in the ACC domain, suggestive of no major transport of particulate lithogenic Fe to our study area. *Ridame and Guieu* [2002] calculated settling velocities in seawater of 0.05 to ~18 m d<sup>-1</sup> for particles from 1 to 20 μm. In 1 year, large particles would thus settle down to the bottom and not reach our study area. However, small particles weathered close to South America and dissolved during the transport to the Greenwich meridian could explain the deep



**Figure 7.** Modeled surface accumulation of dissolved iron ( $\text{nmol L}^{-1}$ ) from 1 November, considering only surface flux processes based on NCAR or INCA dust climatologies, using a 3.5% Fe content and either a (a) 5% Fe solubility or a (b) 10% Fe solubility. Physical advection and diffusion are obtained by an adjusted model configuration (SAFE [Penven *et al.*, 2006]).

DFe concentrations observed in this domain. In deep waters close to the MAR, high values of DFe suggest hydrothermal inputs [Boyle and Jenkins, 2008; Bennett *et al.*, 2008; Tagliabue *et al.*, 2010], and high  $\text{Fe}_{\text{app}}$  concentrations measured at the bottom of S2 and S3 are in accordance with slope currents of AABW observed near the oceanic ridge [Gladyshev *et al.*, 2008].

[28] At station L6 located in the PF, an increase in the surface DFe concentrations was observed with values higher than  $0.2 \text{ nmol L}^{-1}$ . The polar frontal region of the ACC is characterized by a rapid eastward flowing jet. With a velocity of  $25 \text{ cm s}^{-1}$  [Cunningham and Pavic, 2007; Arhan *et al.*, 2002], a mean transit time of 1.3 years with a minimum value of 8 months was calculated. This imprint is thus likely to be seen in surface waters. Besides, active upwelling and eddy diffusion caused by vigorous wind and high deep DFe concentrations ( $0.6\text{--}1.1 \text{ nmol L}^{-1}$ ) are supposed to supply Fe to surface waters of the polar front [Löscher *et al.*, 1997]. Considering the same approach as Löscher *et al.* [1997], we calculated the upwelled and diffusive fluxes. This leads to

a total upward flux of  $1.7 \times 10^{-12} \text{ mol Fe m}^{-2} \text{ s}^{-1}$ , similar to previous values [Löscher *et al.*, 1997; Croot *et al.*, 2004] and largely dominated by the advective flux.

#### 4.1.3. Weddell Sea Gyre (Station S5)

[29] This region was characterized by low DFe concentrations and high  $\text{Fe}_{\text{app}}$  concentrations. The phytoplankton in this area was in a growing stage [Le Moigne *et al.*, 2009], which can explain low values of DFe and high concentrations of biogenic particulate Fe. Horizontal advection of lithogenic particles from the west could also explain the high  $\text{Fe}_{\text{app}}$  values at S5. The Weddell-Scotia confluence (WSC) is indeed the largest productive area in the Southern Ocean [Kahru *et al.*, 2007] because of Fe inputs from the Weddell Sea [Nolting *et al.*, 1991]. Orsi *et al.* [1993] observed the physical WSC signal as far as  $22^\circ\text{E}$ . Finally, icebergs, which we observed in this region, can be a local source of Fe in surface waters [Löscher *et al.*, 1997].

## 4.2. Distribution of SFe and CFe

[30] The study of the different Fe sources is of importance to understand how this essential micronutrient may affect primary production in the Southern Ocean. However, its impact on phytoplankton is related to its bioavailability, and DFe is not as bioavailable as previously thought. A significant part of this pool is indeed present under colloidal form [Wu *et al.*, 2001]. Colloids, in particular when older than 15 days, have been shown to be less available than SFe [Chen and Wang, 2001]. However, open ocean data on these fractions are lacking to improve our understanding of the Fe cycle. The following paragraphs discuss the distribution of SFe and CFe along the BONUS-GoodHope transect and further explores the interactions between those two pools.

[31] No statistically significant correlation between DFe and SFe was observed ( $R^2 = 0.35$ ,  $n = 48$ ,  $p = 0.6$ ; data not shown). Because CFe is calculated using DFe values, auto-correlation prevents us from examining any relationship between DFe and CFe. Different hypotheses, which are not exclusive, can be formulated to explain CFe and SFe distributions.

[32] 1. The first hypothesis is colloidal aggregation and vertical export of CFe in the water column through scavenging.  $\text{Fe}_{\text{app}}$ , DFe, and CFe concentrations observed in the upper 120 m (except at station S1; see below) presented minimum values at the same depths, suggesting that Fe is not simply partitioned between the dissolved and the particulate fractions. The same observation was reported in some studies and was attributed to vertical export of Fe through scavenging [Sedwick *et al.*, 2005; Sarthou *et al.*, 2007]. Here, colloidal aggregation followed by sinking could thus be an explanation for the CFe minimum in surface waters and the increase in CFe concentrations with depth.

[33] 2. The solubility of Fe is related to the concentrations of soluble and colloidal ligands. Liu and Millero [2002] determined a Fe (III) solubility of  $0.5 \text{ nmol L}^{-1}$  in  $0.02 \mu\text{m}$  of filtered seawater and of  $11 \text{ pmol L}^{-1}$  in  $0.7 \text{ mol L}^{-1}$  NaCl ( $S = 36$ ,  $T = 5^\circ\text{C}$ , and  $\text{pH} = 8$ ). They concluded that the difference between these two values was due to the presence of soluble Fe-binding organic ligands in seawater. At station S1, located in the subtropical domain, both CFe and SFe concentrations were high ( $0.2 \text{ nmol L}^{-1}$  at 30 m for CFe,  $0.4\text{--}0.6 \text{ nmol L}^{-1}$  in the upper 1500 m for SFe) compared to the concentrations in the other domains of the section

(<0.07 nmol L<sup>-1</sup> for CFe in surface waters, 0.1–0.4 nmol L<sup>-1</sup> in the upper 1500 m for SFe). The difference between the microbial planktonic communities may explain the difference between the subtropical domain and the other ones. Numerous studies have shown that prokaryotes (heterotrophic bacteria or cyanobacteria) produce soluble organic molecules such as siderophores that bind Fe and make it more available [Reid and Butler, 1991; Wilhelm and Trick, 1994; Butler, 1998; Mawji et al., 2008]. Unfortunately, during the BONUS-GoodHope cruise, no data on heterotrophic bacteria activity were available to assess the hypothesis of a ligand production by these species. However, at station S1, a bloom in a senescent stage dominated by cyanobacteria was observed (J. Ras and H. Claustre, personal communication, 2009). It could induce a high concentration of siderophores. The senescent stage of the bloom also implies that colloidal ligands such as porphyrines or transparent exopolymer particles may have been released during cell lysis or grazing [Rue and Bruland, 1995; Rue and Bruland, 1997; Leppard, 1997]. It could explain the elevated CFe concentrations measured in the surface waters at the S1 station. Another explanation is the advection of water masses enriched with ligands from continental origin through the Agulhas Current, together with Fe (see section 4.1.1).

[34] 3. The third hypothesis involves biological uptake of the CFe and/or SFe with a subsequent exchange between CFe and SFe. Given that SFe concentrations in surface waters are not depleted, whereas CFe are (except at station S1), our results suggest that CFe may be used for uptake by phytoplankton. Possible mechanisms of CFe uptake are biological reduction at the cell surface [Maldonado and Price, 2000, 2001] or direct ingestion [Barbeau et al., 1996]. Some studies have demonstrated the bioavailability of CFe [Kuma et al., 2000; Chen and Wang, 2001], especially freshly precipitated colloids (<15 d [Chen and Wang, 2001]). However, given that DFe in the ACC domain is suspected to come from the Antarctic Peninsula, South America, and/or the South Georgia Islands, and that the transport time is of 1–2 years, colloids would be aged and may not be easily assimilated. An assimilation of SFe followed by a rapid exchange between the colloidal and the soluble iron could occur [Wells, 2003; Morel et al., 2008].

## 5. Conclusion

[35] The study of the Fe speciation along the BONUS-GoodHope section provides information on the distribution and the sources of Fe in the Atlantic sector of the Southern Ocean.

[36] Dust inputs coming from Patagonia and southern Africa (followed by advection of water masses) seem to be an important source explaining the high Fe<sub>app</sub> and DFe concentrations observed in surface waters within the subtropical domain. It is encouraging that despite the great differences between ROMS-SAF<sub>e</sub> and PISCES (in terms of model complexity), their overall conclusions regarding the importance of dust Fe are similar. However, large uncertainties and unknowns persist on the estimation of dust fluxes to the open ocean [Wagener et al., 2008]. The estimation of Fe deposition fluxes in remote areas and of the solubility of these aerosols have to be improved to better constrain this important source of Fe to the open ocean. Inputs of lithogenic Fe transported by

slope currents near the African continental margin is an additional explanation to the high DFe and Fe<sub>app</sub> concentrations observed in the subtropical domain. This process of Fe enrichment is also observed at the bottom of the stations located near the ridge in the ACC domain. Moreover, the ACC domain is characterized by strong currents that can transport Fe from the Drake Passage, the Antarctic Peninsula, and/or the South Georgia Islands. An estimated upwelling flux of  $1.7 \times 10^{-12}$  mol Fe m<sup>-2</sup> s<sup>-1</sup> in the polar frontal region was similar to previous values [Löscher et al., 1997; Croot et al., 2004] and could explain the higher values observed in surface waters of the L6 station. Lateral advection of the WSC, continental shelf sediments or melting icebergs could be the source of Fe<sub>app</sub> south of the SBdy. Hydrothermal inputs could also contribute to some local enrichment of DFe in deep waters.

[37] SFe exhibited the same vertical profiles as samples collected in the Atlantic Ocean [Wu et al., 2001; Bergquist et al., 2007], except in the subtropical domain. Organic ligands released by cyanobacteria and/or advected through the Agulhas Current could explain higher SFe and CFe values observed in this domain. Colloidal aggregation and sinking of CFe, and assimilation of SFe followed by a rapid exchange between the colloidal and the soluble Fe, are also suspected.

[38] More studies on Fe speciation in the soluble and colloidal fractions need to be performed, both in situ and in laboratory process studies, to better constrain the question of the bioavailability of Fe. This would aid further developments of models that aim to include Fe speciation and its effect on bioavailability [Tagliabue et al., 2009b].

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