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Key Points:

- CO₂ response to SAM was different between zones in the south and north
- This difference was associated with regional oceanographic processes
- SAM shift may reverse the negative trend of Southern Ocean CO₂ uptake

Supporting Information:

Text S1, Figures S1 and S2, and Table S1

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Response of sea surface fugacity of CO₂ to the SAM shift south of Tasmania: Regional differences

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Abstract Using observational data collected south of Tasmania during 14 austral summer cruises during 1993–2011, we examined the response of sea surface fugacity of carbon dioxide (fCO_2) to the Southern Annular Mode (SAM) shift, which occurred around 2000. In the southern part of the Southern Ocean (SO) or the Polar Zone (PZ) and the Polar Frontal Zone (PFZ), fCO_2 increased faster at the sea surface than in the atmosphere before the SAM shift, but not after the shift. In the northern part of the SO or the Subantarctic Zone (SAZ), however, surface fCO_2 increased faster than atmospheric fCO_2 both before and after the shift. The SAM shift had an important influence on the surface fCO_2 trend in the PZ and PFZ but not in the SAZ, which we attribute to differences in regional oceanographic processes (upwelling versus nonupwelling). The SAM shift may have reversed the negative trend of SO CO₂ uptake.

1. Introduction

The Southern Ocean (SO, south of 44°S) is a key region for the uptake of anthropogenic carbon dioxide (CO₂), accounting for about one third of the total CO₂ uptake by the global ocean (0.7 Pg C yr⁻¹, 1 Pg = 10^{15} g) [*Gruber et al.*, 2009; *Mikaloff Fletcher et al.*, 2006]. As such, it plays a significant role in slowing the accumulation of CO₂ in the atmosphere and in regulating the Earth's climate system [*Lovenduski*, 2012; *Takahashi et al.*, 2012]. As a result, increasing attention is being paid to CO₂ uptake by the SO, and more efforts have been made to evaluate changes in it via model and observational studies [*Fay and McKinley*, 2013; *Fay et al.*, 2014; *Landschützer et al.*, 2014; *Le Quéré et al.*, 2007; *Lenton and Matear*, 2007; *Lenton et al.*, 2012, 2013; *Lovenduski et al.*, 2007, 2015; *Majkut et al.*, 2014; *Metzl*, 2009; *Takahashi et al.*, 2009, 2012; *Yoshikawa-Inoue and Ishii*, 2005].

However, there are discrepancies among published results. Some model studies have showed that a recent reduction in the SO CO₂ uptake was probably associated with strengthened upwelling of CO₂-rich deep waters during a more positive phase of the Southern Annular Mode (SAM) [*Hauck et al.*, 2013; *Lenton and Matear*, 2007; *Lovenduski et al.*, 2007]. In sharp contrast, *Majkut et al.* [2014] reported an increased CO₂ uptake during a positive SAM trend with an analysis based on a Markov Chain Monte Carlo method. Also, *Lenton et al.* [2013] found discrepancies between the trends in SO CO₂ uptake obtained by ocean biogeochemical models (an increase) and by atmospheric inversions (little change). With increasing efforts in the SO observations, the trend in SO CO₂ uptake was recently evaluated by examining the change of CO₂ fugacity (*f*CO₂) measured at the sea surface on a regional or basin scale [*Fay et al.*, 2014; *Metzl*, 2009], since atmospheric CO₂ was almost homogeneous across the globe and usually increased steadily with time [*Takahashi et al.*, 2014]. However, conflicting results were also obtained even for analyses based on the same CO₂ data set [*Fay and McKinley*, 2013; *Fay et al.*, 2014; *Lenton et al.*, 2012; *Takahashi et al.*, 2012], probably associated with methodological choice such as how data are grouped or how regions are defined [*Fay et al.*, 2014].

Although progress has been made in recent years, it is still a significant challenge to evaluate the trend in SO fCO₂ on the basin scale via synthesis of observational data [*Fay et al.*, 2014; *Kouketsu and Murata*, 2014; *Lenton et al.*, 2013; *Lovenduski et al.*, 2015; *Takahashi et al.*, 2012]. First, the SO is one of the most under-sampled regions in the globe [*Fay and McKinley*, 2013; *Fay et al.*, 2014; *Takahashi et al.*, 2009]. Second, the SO hosts strong spatiotemporal variability and the SO fCO₂ change is not zonally uniform, e.g., between the Atlantic and the Pacific sectors [*Lenton et al.*, 2012]. Furthermore, fCO₂ data in the SO have commonly been

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collected in the austral summer and are relatively sparser in the austral winter because of hostile weather conditions [*Lenton et al.*, 2013; *Takahashi et al.*, 2012].

The SAM is the dominant mode of climate variability in the extratropical Southern Hemisphere [*Hall and Visbeck*, 2002; *Lovenduski*, 2012]. A positive SAM trend is correlated with enhancements in westerly winds, northward Ekman transport and meridional overturning, and anomalous upwelling in the region south of the Antarctic Circumpolar Current [*Hall and Visbeck*, 2002; *Lovenduski*, 2012]. Although the SAM index increased after the mid-1980s [*Marshall*, 2003], a decrease was observed from 2000 [*Johnston and Gabric*, 2011]. That is, a shift in the SAM trend occurred around 2000, which may have altered the trend in SO fCO_2 , as the Pacific Decadal Oscillation shift did in the equatorial Pacific [*Takahashi et al.*, 2003]. Analyzing the SAM shift, studies based on the entire SO observational data indicate that the SO CO_2 uptake has strengthened from the early 2000s [*Fay and McKinley*, 2013; *Landschützer et al.*, 2014] or from 2007 [*Fay et al.*, 2014]. This difference over time could have been associated with the challenge in evaluating the trend of SO fCO_2 on the basin scale as mentioned above. Thus, at present, it probably is better to evaluate the change of oceanic fCO_2 within a small region (e.g., the meridional transect) that has well-defined oceanographic features and is rich in observational data.

More important, since the influence of the SAM on CO₂ uptake is associated with upwelling [*Le Quéré et al.*, 2007; *Lovenduski et al.*, 2007; *Metzl*, 2009] and the SO undergoes upwelling in the south and subduction in the north [*Anderson et al.*, 2009], there will be regional differences in the response of fCO₂ to the SAM, which, however, has not been highlighted sufficiently before [*Fay and McKinley*, 2013].

In this work, we examine the response of sea surface fCO_2 to the SAM shift and highlight its regional differences among circumpolar zones, using transect data collected south of Tasmania in the austral summer.

2. Data Sources and Methods

We used data collected during 14 austral summer cruises (December–March) from two nearby transects (Transects SR03 and Re, Figure 1) south of Tasmania to examine changes in oceanic *f*CO₂. Detailed information about the data can be found in Table S1 in the supporting information. Sea surface *f*CO₂, temperature (SST), salinity (SSS), and barometric pressure data were extracted from the Surface Ocean CO₂ Atlas (SOCAT) (http://www.socat.info/) [*Bakker et al.*, 2014]. Atmospheric CO₂ data in January were from the GCO (Cape Grim, Tasmania) atmospheric CO₂ measurement station (Figure 1a, ftp://aftp.cmdl.noaa.gov/data/trace_gases/co2/flask/). The CO₂ mole fraction was converted to *f*CO₂ as in *Jiang et al.* [2008].

To reduce uncertainty in the trend analysis due to spatial variability and to examine regional differences in fCO_2 changes, we divided the study area from north to south into three zones according to the positions of the main circumpolar fronts (Figure 1), i.e., the Subantarctic Zone (SAZ) between the Subtropical Front (STF) and the Subantarctic Front (SAF), the Polar Frontal Zone (PFZ) between the SAF and the Polar Front (PF), the Polar Zone (PZ) between the PF and 62°S [*Yoshikawa-Inoue and Ishii*, 2005]. The positions of the STF, SAF, and PF were determined based mainly on SSS gradients [*Chaigneau and Morrow*, 2002], since surface data were relatively easily available, and fCO_2 changes at the surface were focused on [*Midorikawa et al.*, 2012; *Nakaoka et al.*, 2009; *Yoshikawa-Inoue and Ishii*, 2005]. The specific position of each front was shown in Table S1 and Figure S1. In addition, to reduce errors due to different sampling intervals, for each parameter (including SST, SSS, and sea surface fCO_2) we first binned all the data points into their respective 0.02° latitudinal bands, then calculated the average for each band, and finally took an average at each zone as in *Xue et al.* [2014]. Linear regression analyses for sea surface fCO_2 and other related parameters were performed using the mean value of each parameter within each zone. The trend with *p* value < 0.1 was regarded as significant statistically at 90% confidence level due to small sample numbers.

3. Results and Discussion

3.1. Sea Surface fCO₂ Changes Before and After the SAM Shift

It seems that sea surface fCO_2 responded noticeably to the SAM shift (Figures 2 and 3). The SAM index showed a significant increase during 1993–2000 and a general decrease during 2000–2011 (though not significant), i.e., a shift in the SAM trend occurred around 2000 (Figure 2). Correspondingly, sea surface

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Figure 1. (a) Sigma-theta (kg m⁻³) (shaded and solid contours) and salinity (white dashed contours) profiles along Transect SR03 south of Tasmania in January 1994 (Data are from ftp://data.nodc.noaa.gov/pub/data.nodc/woce/version3.0/), the study area (the insert), and (b) a schematic depiction of the influence of the SAM shift on the CO₂ uptake in the southern SO (upwelling-affected zone) based on *Lovenduski and Gruber* [2005]. In Figure 1a, positions of main circumpolar fronts (STF, SAF, and PF) and zones (SAZ, PFZ, and PZ) together with the locations of Subantarctic Mode Water (SAMW), Antarctic Intermediate Water (AAIW), and Upper Circumpolar Deep Water (UCDW) were shown. The schematic of Ekman upwelling and downwelling in the SO based on annual wind stress curl [*Trull et al.*, 2001] was also shown. In the insert, the black and red lines represent Transects SR03 and Re; the red triangle showed the GCO (Cape Grim, Tasmania) atmospheric CO₂ measurement station (40.683°S, 144.690°E). In Figure 1b, during a positive SAM trend, increased westerly winds induced strengthened upwelling, which enhanced upward transport of CO₂- and nutrient-rich subsurface waters and thus increased chlorophyll *a* (Chl *a*) levels. During this period, CO₂ uptake decreased due to faster *f*CO₂ increases at the sea surface than in the atmosphere (i.e., decrease in the absolute value of air-sea *f*CO₂ gradient, $|\Delta fCO_2|$). During a negative SAM trend, in contrast, decreased westerly winds induced weakened upwelling, which suppressed upward transport of CO₂- and nutrient-rich subsurface waters and thus surface waters and thus decreased westerly winds induced the sea surface than in the atmosphere (i.e., decrease in the absolute value of air-sea *f*CO₂ gradient, $|\Delta fCO_2|$). During a negative SAM trend, in contrast, decreased westerly winds induced weakened upwelling, which suppressed upward transport of CO₂- and nutrient-rich subsurface waters and thus atmosphere (i.e., increase in $|\Delta fCO_2|$).

 fCO_2 increased faster before the SAM shift (i.e., in 1993–2000) than after the shift (i.e., in 2000–2011) (Figure 3 and Table 1). Especially in the southern part of the SO or the PZ and PFZ, before the SAM shift, sea surface fCO_2 increased rapidly at rates of $4.48 \pm 1.04 \,\mu$ atm yr⁻¹ and $5.20 \pm 2.55 \,\mu$ atm yr⁻¹, respectively, faster than the atmospheric fCO_2 growth rate of $1.79 \pm 0.10 \,\mu$ atm yr⁻¹. In contrast, after the shift, sea surface fCO_2 did not increase significantly. In the northern part of the SO or the SAZ, however, surface fCO_2 increased faster than the atmospheric fCO_2 both before and after the SAM shift, with rates of $3.05 \pm 1.06 \,\mu$ atm yr⁻¹ and $2.33 \pm 0.72 \,\mu$ atm yr⁻¹, respectively. It appears that the proposed mechanism of increased upwelling of CO₂-rich deep waters during a positive SAM trend in the early SO carbon study [*Lenton and Matear*, 2007; *Lovenduski et al.*, 2007] may be used to explain the observed higher rates of sea surface fCO_2 increase before the SAM shift than after it. However, oceanic fCO_2 is also affected by temperature thermodynamically, in addition to processes that change CO₂ chemistry such as mixing and biological activity. Therefore, using the formula of *Takahashi et al.* [1993], we normalized sea surface fCO_2 to the average temperatures of each zone (4.15°C, 8.84°C, and 12.02°C in the PZ, PFZ, and SAZ), nfCO₂, and examined the trend in nfCO₂, which removes the temperature effect and reflects CO₂ chemistry changes [*Xue et al.*, 2012].



Figure 2. SAM index in January calculated by *Marshall* [2003]. The empty circle, black filled circle, and gray filled circle denoted the data during the periods of 1957–1992, 1993–2000, and 2000–2011. Linear regression analyses were performed for data during 1993–2000 and 2000–2011, respectively.

In the PZ and the PFZ, surface nfCO₂ still increased quickly before the SAM shift (though not significantly in the PFZ), but almost had no changes after the shift (Figure 3). This suggests that the fCO₂ increase in these two zones before the SAM shift was primarily induced by an increased CO₂ concentration, most likely associated with enhanced upwelling [Lenton and Matear, 2007; Lovenduski et al., 2007; Metzl, 2009]. Also, because of the increasing nfCO₂, we suggest that enhanced biological production (chlorophyll a) due to increased iron supply by upwelling during a positive SAM trend [Lovenduski and Gruber, 2005] only partly counteracted the CO₂ increase even in



Figure 3. (a–c) Temporal changes of sea surface fCO_2 , (d–f) temperature normalized fCO_2 (nfCO₂), and (g–i) air-sea fCO_2 gradient (ΔfCO_2) in the PZ, PFZ, and SAZ. Linear regression analyses were performed for the period 1993–2000 and 2000–2011, respectively. During the regression analysis, red data were not considered and data collected in December 2000 were grouped into the period 2000–2011, rather than the period 1993–2000. In the figure, *p* values were presented. Slopes can be found in Table 1. The bars showed 1 standard deviation. In Figure 3a, atmospheric fCO_2 data in January collected at the GCO (Figure 1) were also presented.

Table 1. Change Rates of Air fCO_2 , Seawater fCO_2 , Temperature Normalized fCO_2 ($nfCO_2$), and Air-Sea fCO_2 Gradient ($\Delta fCO_2 = fCO_{2sea} - fCO_{2air}$) in the PZ, PFZ, and SAZ during Different Periods^a

Period	Zones	Air <i>f</i> CO ₂	Seawater fCO ₂	nfCO ₂	$\Delta f CO_2$
1993–1999	PZ	$1.79 \pm 0.10^{*}$	$4.48 \pm 1.04^{*}$	$2.48 \pm 0.98^{*}$	$2.70 \pm 0.97^{*}$
	PFZ		$5.20 \pm 2.55^*$	5.18 ± 3.72	3.43 ± 2.56
	SAZ		3.05 ± 1.06*	-0.59 ± 3.80	1.25 ± 1.03
2000-2011	PZ	$1.92 \pm 0.03^{*}$	0.92 ± 0.51	-0.18 ± 1.50	$-1.12 \pm 0.52^{*}$
	PFZ		0.72 ± 0.82	1.27 ± 2.25	-1.31 ± 0.81
	SAZ		$2.33 \pm 0.72^{*}$	4.94 ± 2.31*	0.30 ± 0.81
1993-2011	PZ	$1.85 \pm 0.02^*$	$1.31 \pm 0.35^*$	2.16 ± 0.86*	-0.55 ± 0.36
	PFZ		$2.09 \pm 0.58^{*}$	3.05 ± 0.96*	0.23 ± 0.60
	SAZ		$1.80 \pm 0.43^{*}$	1.81 ± 1.03*	-0.05 ± 0.43

^aUnits are in μ atm yr⁻¹.

*p value < 0.1 (statistically significant at 90% confidence level).

the austral summer, a productive season [*Trull et al.*, 2001]. This argument is consistent with the inference of *Lovenduski and Gruber* [2005] and some model results [*Hauck et al.*, 2013; *Lenton and Matear*, 2007]. In the SAZ, nfCO₂ had no trend before the SAM shift, while after the shift the rate of nfCO₂ increase was greatly enhanced to $4.94 \pm 2.31 \,\mu$ atm yr⁻¹ (Figure 3), suggesting the insignificant influence of the SAM shift on nfCO₂ in this zone. Also, it indicates that the high fCO₂ increase rate in the SAZ before the SAM shift was not due to net changes in CO₂ chemistry but was primarily due to temperature increase (warming, Figure S2). In contrast, the high rate of fCO₂ or nfCO₂ increase in the SAZ after the SAM shift was caused by an increase in CO₂ concentration, although cooling partly offsets the fCO₂ increase (Table 1 and Figure 3). However, we are not clear about the process driving the increase in CO₂ concentration after the SAM shift; the SAZ might have been affected by other climate phenomena such as El Niño–Southern Oscillation likely by modulating the transport of the East Australian Current extension [*Ayers and Strutton*, 2013]. Overall, the SAM shift had a clear influence on the trend of surface *f*CO₂ in the PZ and the PFZ but not in the SAZ.

The different response of fCO_2 to the SAM shift between the PZ and PFZ in the south and the SAZ in the north was probably related to regional properties (Figure 1). A positive SAM trend was associated with enhanced westerly winds in the PZ and the PFZ indicating an increased upwelling, and in contrast, it was related to an enhanced easterly wind in the SAZ implying an increased downwelling [*Lovenduski and Gruber*, 2005; *Ayers and Strutton*, 2013]. This probably makes the response of sea surface fCO_2 to the SAM in the PZ and the PFZ different from that in the SAZ.

The PZ was greatly affected by upwelling of upper circumpolar deep water (Figure 1a) [Trull et al., 2001; Anderson et al., 2009], although the typical upwelling feature of high salinity at the surface was masked by the northward transport of less saline waters from the seasonal sea ice zone [Popp et al., 1999]. Thus, sea surface fCO₂ or nfCO₂ increase rate faster than that of atmospheric CO₂ during a positive SAM trend (Figure 3a and Table 1) was most likely caused by enhanced upwelling driven by increased westerly winds during this period [Hall and Visbeck, 2002; Lenton and Matear, 2007; Lovenduski et al., 2007; Metzl, 2009] (Figure 1b). In contrast, after the SAM shift sea surface fCO2 or nfCO2 increase was not significant (Figure 3a and Table 1), due possibly to weakened upwelling. The different responses of oceanic fCO_2 to the SAM shift as well as the steady increase in atmospheric CO₂ during these two periods (Figure 3) indicate that on relatively short timescales, changes in fCO2 were more sensitive to upwelling strength than to increase in anthropogenic CO2, although oceanic CO2 increase was essentially driven by increasing anthropogenic CO₂ on a long-term timescale [e.g., Khatiwala et al., 2013; Xue et al., 2014]. This also should be further examined in other upwelling regions such as the coastal upwelling system where upwelling strength was enhanced due to global warming [Bakun, 1990]. Although the PFZ was not affected by direct upwelling of upper circumpolar deep water as strongly as the PZ, upwelled water moved northward at the surface forced by prevailing westerlies [Anderson et al., 2009], also influencing surface waters (Figure 1a). Thus, the sea surface fCO_2 response to the SAM shift in the PFZ was similar (to) but not as strong as that in the PZ. In the SAZ, where subduction occurred and mode waters formed [Anderson et al., 2009], surface water in this zone might not have been affected by upwelled waters moving northward, since after passing the PFZ these upwelled waters were subducted along a relatively deep isopycnal surface and may have not accumulated at the surface (Figure 1).

Our regional analysis shows that sea surface fCO_2 increases in the PZ and the PFZ (corresponding to the ice biome and subpolar seasonally stratified biome, respectively [*Fay and McKinley*, 2013]) greatly slowed after the SAM shift (Table 1 and Figure 3). This result is generally consistent with that by basin-scale studies [*Fay and McKinley*, 2013; *Fay et al.*, 2014; *Landschützer et al.*, 2014], despite some differences in the time when the fCO_2 trend began to slow. Nonetheless, our study found that the sea surface fCO_2 trend in the SAZ (corresponding to the subtropical seasonally stratified biome [*Fay and McKinley*, 2013]) was not sensitive to the SAM shift, differing from previous results [*Fay and McKinley*, 2013]. Previous work found that in the SAZ from the 1990s to 2009–2010 during a weak negative SAM trend, sea surface fCO_2 increased slower than atmospheric CO_2 , while in our regional study during 1993–2011 fCO_2 increase at the sea surface was not distinct from that in the atmosphere (Table 1). This difference is probably related to regional differences or to data scarcity in the SO basin [*Fay et al.*, 2014]. In addition, *Fay and McKinley* [2013] found that cooling alleviated the increase in sea surface fCO_2 for the period from the 1980s to 2007 or later in the entire SO, and we also found similar phenomena during 1993–2011 in the PZ and PFZ, rather than in the SAZ, as revealed by change rates of fCO_2 and $nfCO_2$ (Table 1).

3.2. Potential Impact on Southern Ocean CO₂ Uptake

The SAM shift also impacted air-sea fCO_2 gradient ($\Delta fCO_2 = fCO_{2sea} - fCO_{2air}$, Figure 3), the driving force of air-sea CO_2 exchange, and thus the SO CO_2 uptake (Figure S2). In the PZ, before the SAM shift, ΔfCO_2 showed a positive trend (i.e., decrease in the absolute value of ΔfCO_2 , $|\Delta fCO_2|$), and a negative trend after it (i.e., increase in $|\Delta fCO_2|$, Figure 3 and Table 1). Response of ΔfCO_2 to the SAM shift in the PFZ was similar (to) but not as strong as that in the PZ. In the SAZ, ΔfCO_2 increased (though not significantly) before the SAM shift, but almost had no changes after the shift (Figure 3). From the aspect of ΔfCO_2 , as done by a number of studies examining the trend of CO_2 uptake [*Fay and McKinley*, 2013; *Fay et al.*, 2014; *Lenton et al.*, 2012; *Metzl*, 2009; *Takahashi et al.*, 2012], CO₂ uptake will decline before the SAM shift and will increase after it in the southern SO (PZ and PFZ, i.e., upwelling-affected zone).

While oceanic CO₂ uptake was also influenced by gas transfer velocity, a function of wind speed, its role was relatively minor. The calculated air-sea CO₂ flux (see flux calculation in the supporting information) showed that the PZ CO₂ uptake decreased before the SAM shift, despite that wind speed increased during this period, and it increased after the shift (Figure S2). It indicated that air-sea fCO_2 gradient (ΔfCO_2) dominated the CO₂ uptake in the SO, consistent with the result of *Lovenduski et al.* [2015]. Overall, during the austral summer, there was a decrease in CO₂ uptake in the southern SO (upwelling-affected zone) before the SAM shift and an increase after it (Figure S2), and the strength of the CO₂ sink in the upwelling-affected zone was closely related to the SAM on short timescales (Figure 1b). Our result about variability in CO₂ uptake just in the southern SO was in general agreement with previous results obtained in the entire SO [*Fay et al.*, 2014; *Hauck et al.*, 2013; *Lenton and Matear*, 2007; *Lovenduski et al.*, 2007; *Metzl*, 2009]. The entire SO CO₂ sink may have increased after the SAM shift around 2000, and more observational efforts in the future are required to confirm this.

4. Conclusions

Although there were uncertainties in the fCO_2 trend due to short observation timescales, it indicates that the SAM shift had an important influence on the sea surface fCO_2 trend in the PZ and the PFZ, but not in the SAZ, because of regional oceanographic processes. Before the SAM shift, high rates of fCO_2 increase in the PZ and the PFZ were probably associated with strengthened upwelling of CO_2 -rich deep waters, and in contrast, the high fCO_2 increase rate in the SAZ was due to surface warming. Thus, response of oceanic fCO_2 and thus CO_2 uptake to climate change was associated with local oceanographic processes (e.g., upwelling versus nonupwelling), which should be emphasized in the future. This paper also provides observational evidence that the variability of the CO_2 sink in the southern SO (upwelling-affected zone) was closely related to the SAM, and the SAM shift around 2000 may have reversed the negative trend of SO CO_2 uptake.

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