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# Nitrous oxide research progress in polar and sub-polar oceans

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**Abstract** $N_2O$ gas depletes ozone and has a powerful greenhouse effect. Oceans are among the most important $N_2O$ sources and have been the subject of extensive studies. Polar oceans are important regions for deep water formation and global-scale thermohaline circulation. Therefore, these water bodies play an important role in the $N_2O$ budget, however, these regions were not well studied. This review of previously published studies and data on polar oceans, including both the Arctic Ocean and Southern Ocean, describes the distribution pattern of $N_2O$ and possible regulating mechanism of these distribution patterns and shows that the Arctic Ocean and Southern Ocean both represent source and sink regions, suggesting that the source/sink characteristics of the Arctic and Southern oceans and their strengths need further study. Questions related to $N_2O$ circulation in polar oceans were proposed, and future work is suggested.

**Keywords** **nitrous oxide, source/sink, Arctic Ocean, Southern Ocean, Prydz Bay**

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## 1 Introduction

$N_2O$ is a trace gas that contributes ~298 times more to the greenhouse effect than $CO_2$ on a per-molecule basis (https://www.epa.gov/ghgreporting/ghg-reporting-program-data-sets); moreover, it has become the dominant contributor to ozone depletion since chlorofluorocarbon (CFC) emissions were eliminated by the Montreal Protocol (Ravishankara et al., 2009). Therefore, it is of wide concern, and the Intergovernmental Panel on Climate Change (IPCC) continues to update information on its contribution to global warming, the global budget, etc. (IPCC, 2001, 2007).

Oceans are considered one of the most important sources of $N_2O$ (Nevison et al., 1995; Seitzinger and Kroeze, 1998; Suntharalingam and Sarmiento, 2000; Freing et al., 2012) and contribute approximately 4 Tg $N{\cdot}a^{-1}$. Previous studies showed that the Atlantic Ocean is a $N_2O$ source (Walter et al., 2006), although its contributions likely have been underestimated. The East Tropical Pacific Ocean and the Arabian Sea have been reported as $N_2O$ production hotspots, where the surface water has high $N_2O$ concentrations and the subsurface layer shows the lowest $N_2O$ concentrations (Cohen, 1978; Naqvi and Noronha, 1991). More recent following studies elucidated that the significant $N_2O$ supersaturation of 12244% at the ETSP (Arevalo-Martínez et al., 2015) and fast $N_2O$ turnover rate of about 20 times at that of outgassing rate of the ETNP (Babbin et al., 2015). In addition to these regions, the Southern Ocean is also predicted to be a significant source of atmospheric $N_2O$ due to possible upwelling in the high-latitude Southern Ocean in $N_2O$-rich Circumpolar Deep Water (CDW) (Bouwman et al., 1995). However, relatively limited surface water $N_2O$ data are available for

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the Southern Ocean (Rees et al., 1997; Zhan et al., 2015a, 2015b, 2018), and the information is insufficient to elucidate the source/sink characteristics and constrain the budget of the Southern Ocean. Although a few studies on the $N_2O$ characteristics of the Arctic Ocean have been carried out (Zhan et al., 2015a, 2015b, 2016; Zhang et al., 2015), this water body has long been ignored in terms of $N_2O$ source/sink characteristics since it is usually covered with sea ice; however, with the retreat of sea ice, its role in the global oceanic budget should be considered. Moreover, significant denitrification may occur along the Arctic continental shelf, which may also be a significant source of atmospheric $N_2O$. Therefore, the role the Arctic Ocean in the $N_2O$ budget must be considered. Although the $N_2O$ characteristics in polar and sub-polar oceans have been less frequently studied, these regions are important locations for thermohaline circulations, where upwelling water approaches the surface and deep water forms; moreover, some of these regions are also considered important $N_2O$ sources. To provide a better understanding of the $N_2O$ research progress in polar and sub-polar oceans, this study reviewed the literature focused on these regions and proposed directions for further investigations.

## 2 Data

The data shown in this work were obtained during the Chinese National Arctic and Antarctic expeditions. Data for the Arctic and sub-Arctic oceans were obtained during the 4th and 5th Chinese National Arctic Research expeditions (CHINAREs-Arctic), which were carried out during 2010 and 2012, respectively, and data for Southern Ocean were obtained during the 27th and 28th Chinese National Antarctic Research expeditions (CHINAREs), which were carried out between 2010 and 2012. The sampling stations are shown in Figure 1. The study area of the Arctic and sub-Arctic oceans includes the Bering Sea, Chukchi Sea, Beaufort Sea and Nordic Sea, and the study area of the Southern Ocean includes the Prydz Bay and the Antarctic Peninsula.

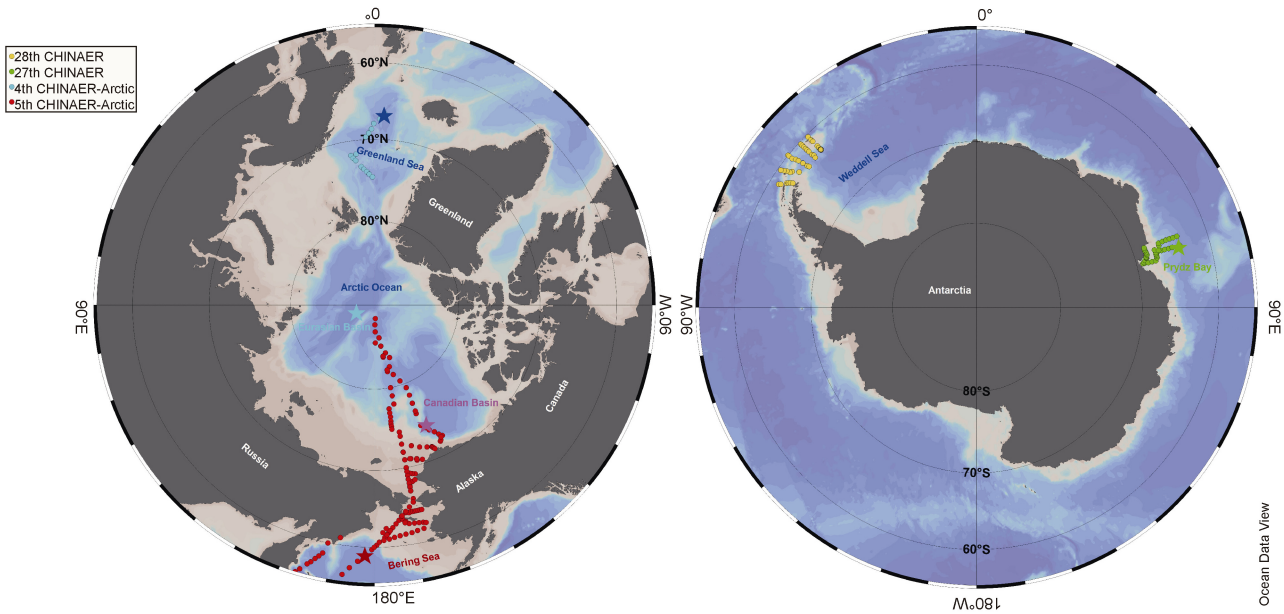


**Figure 1** The air-sea fluxes of $N_2O$ in polar and sub-polar oceans.

## 3 $N_2O$ Distribution in polar and sub-polar oceans

### 3.1 $N_2O$ in the Arctic and sub-Arctic oceans

#### 3.1.1 Arctic continental shelf

In the Arctic Ocean, studies have been conducted on $N_2O$ over continental shelves. Several studies about the $N_2O$ distribution, production mechanism and source/sink characteristics have been carried out along the Bering/Chukchi Sea continental shelf. Hirota et al. (2009) investigated the Bering Sea and Chukchi Sea and used nutrient stoichiometry and isotope data to reveal the mechanism of $N_2O$ production. Their results showed that continental shelf sediment may be a source of the $N_2O$ supersaturation observed in the Bering/Chukchi sea continental shelf water and denitrification in the sediment may be the main production mechanism of $N_2O$. Further studies have reported the role of the Bering/Chukchi sea continental shelf as a $N_2O$ sources (Zhang et al., 2015; Fenwick et al., 2017; Wu et al., 2017) and identified similar saturation states in these regions, suggesting that the supersaturation of $N_2O$ in the Bering Sea and Chukchi Sea is a persistent source of atmospheric $N_2O$ during summer. The melting ice edge near the continental shelf is oversaturated with

$N_2O$ (Kitidis et al., 2010). Two studies have focused on the Northwest Passage of the Arctic Ocean (Kitidis et al., 2010; Fenwick et al., 2017), and in the same passage of the Canadian Arctic Archipelago, degrees of $N_2O$ saturation of 179% and 110% were observed; moreover, denitrification along the western Arctic shelf was believed to be the source of $N_2O$ in the water column (Fenwick et al., 2017).

Thus, the Arctic continental shelf may contain important locations for $N_2O$ production (Hirota et al., 2009). Since the continental shelf of the Arctic area accounts for 20% of the continental shelf area worldwide, it is an active site of denitrification (Chang and Devol, 2009). Therefore, the Arctic continental shelf may be an important $N_2O$ source, and further investigation is required.

### 3.1.2 Arctic and sub-Arctic oceans

The distribution pattern of $N_2O$ in the Arctic Ocean is shown in Figure 2. The black dots are data observed in the Canadian Basin (CB), and the white dots are data obtained in the Eurasian Basin (EB). The data were obtained during the 4th and 5th CHINAREs-Arctic.

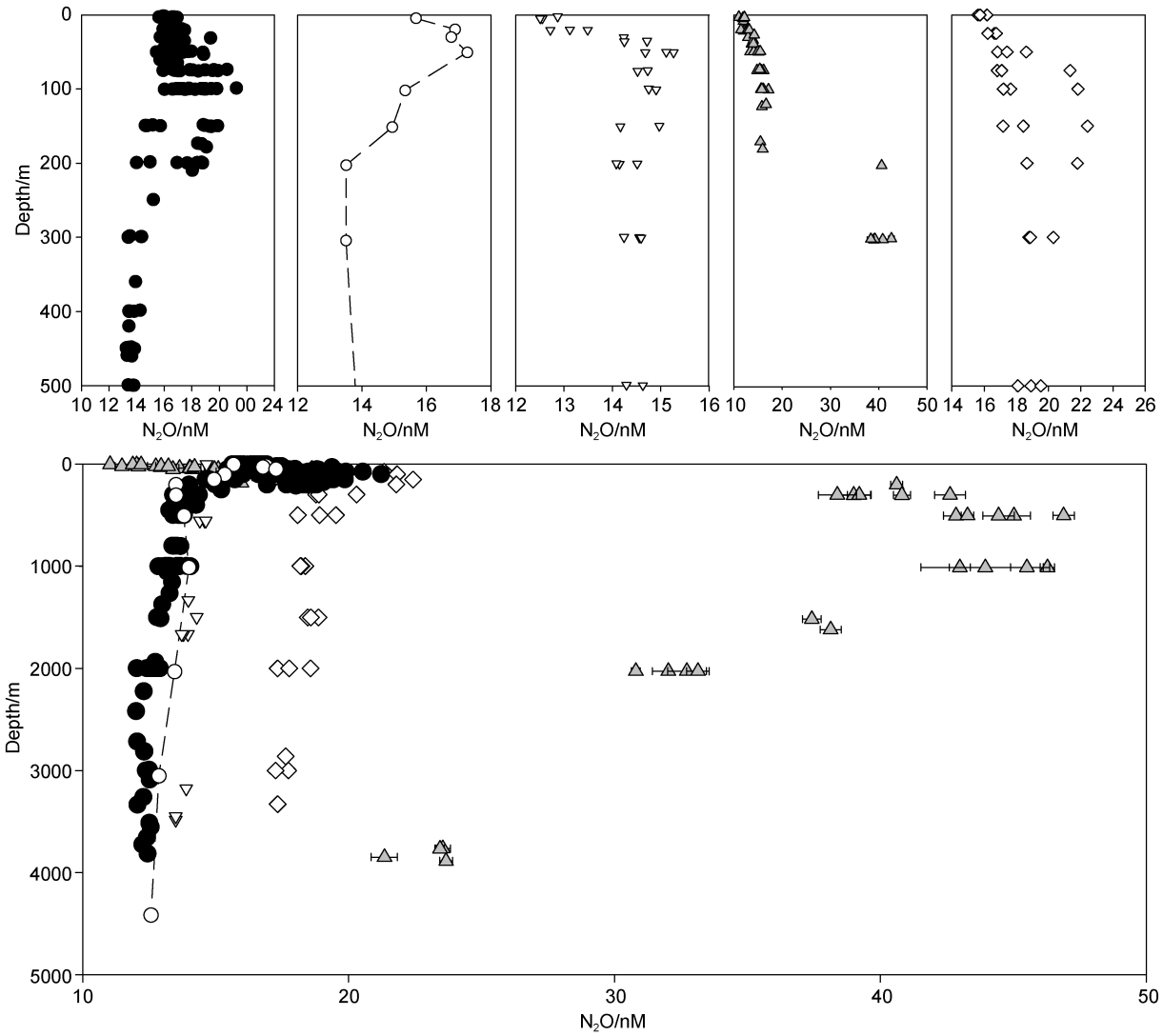


**Figure 2** Vertical distribution of the $N_2O$ concentration in the Canadian Basin (black dots), Eurasian Basin (white circles), Greenland Sea (white triangles), Southern Ocean (north of the Prydz Bay, diamonds) and Bering Sea (gray triangles).

As shown in Figure 2, the $N_2O$ concentrations in the upper 300 m of the two basins are similar. The maximum concentration of ~18 nmol·L$^{-1}$ was observed between 50 m and 100 m, and it declines with depth to ~13.5 nmol·L$^{-1}$ at 300 m. The surface water is well-oxygenated, and nitrification is prohibited by light (Ward, 2008); therefore, the production of $N_2O$ in the surface layer was dominated by the air-sea-ice interaction. The surface water $N_2O$ concentrations are in near-equilibrium with that in the atmosphere. Surface water $N_2O$ undersaturation was observed in the Arctic Ocean, and it had a minimum saturation of approximately 80% (Kitidis et al., 2010;

Zhang et al., 2015). The undersaturation in the Arctic Ocean may be a result of melting ice. The first sea ice $N_2O$ measurement shows that sea ice contains only a low concentration (~6 nmol·kg$^{-1}$) of $N_2O$ (Randall et al., 2012), which will dilute the $N_2O$ in the surface water during the ice melting process. In general, 2–3 weeks is required for the surface water to reach equilibrium again (Butler et al., 1989), which results in surface water undersaturation. The subsurface layer $N_2O$ maximum is generally associated with nitrification that occurs during particulate matter remineralization. The productivity in the surface water of the central Arctic Ocean is limited (Anderson et al., 2003); therefore, sinking particulate matter should be limited and the local production of and contribution to the $N_2O$ maximum should also be limited. Rather, the $N_2O$ maximum should be the result of advection. Zhang et al. (2015) suggests that the $N_2O$ maximum observed at 100 m in the CB is Pacific-origin water (Zhang et al., 2015), and it can still be observed in the EB, where it rises to approximately 50 m. The water temperature increases at 300 m, below which Atlantic-origin water is located.

The $N_2O$ concentrations in the CB and EB are similar at between 300 m and 1000 m, and the average value is approximately 13.5 nmol·L$^{-1}$. The similarity in $N_2O$ concentrations in these two basins at this depth range is because this layer of water originates from the Atlantic, and it descended to this depth and experienced a limited accumulation of $N_2O$ during its transportation (Zhan et al., 2015a).

The difference in $N_2O$ concentrations between the two basins increases with depth below 1000 m, with the largest difference observed between 2000 m and 3000 m. Then, the difference between the two basins decreases again below 3000 m. This phenomenon may be explained by the presence of the Lomonosov Ridge. The CB and EB are separated by the Lomonosov Ridge, which has a depth of ~1500 m and acts as a barrier to prevent deep Atlantic water from flowing into the CB below 1500 m. Therefore, the CB deep water below 1500 m may retain a preindustrial record (Zhan et al., 2015a; Fenwick et al., 2017). Below 3000 m, the $N_2O$ concentrations in the EB and CB are similar, suggesting that the deep bottom water in the EB may be of a similar age as that in the CB.

### 3.2 $N_2O$ and its source/sink characteristics in the Southern Ocean

Several studies have focused on the $N_2O$ in the Southern Ocean. The $N_2O$ concentration in the surface water is close to equilibrium, while that in the surface water was undersaturated. Rees et al. (1997) first observed $N_2O$ undersaturation of approximately 90% in the Bellingshausen Sea; Boontanon et al. (2010) reported $N_2O$ undersaturation of approximately 94% south of 60°S along 140°E; and a similar phenomenon was also observed near Prydz Bay (Zhan and Chen, 2009; Zhan et al., 2015b, 2017). Grefe et al. (2017) also reported 96.5% saturation near the ice edge at the polar front near the Weddell Sea. $N_2O$ undersaturation may also be caused by water dilution from sea ice and glacier melt. This melt water input and solar irradiance warm the stratified Antarctic surface water, which is further transported northward and results in a large area of near-equilibrium surface water. Farías et al. (2014) also observed $N_2O$ undersaturation and suggested that $N_2O$ may be consumed by nitrogen-fixing organisms. All these previous results show that the weak undersaturation of $N_2O$ may be common in the high latitude Southern Ocean.

In the subsurface layer, a $N_2O$ maximum can be observed in the Southern Ocean as well as Prydz Bay (white diamonds). The maximum $N_2O$ concentration in Prydz Bay is approximately 22.4 nmol·L$^{-1}$ as shown in Figure 2 (Zhan et al., 2015b). A similar maximum $N_2O$ concentration can be observed near the Antarctic Peninsula (Zhan et al., 2018), and it corresponds to the core of the upper CDW, which rises to approximately 200 m (Zhan et al., 2015b). The CDW is an "old" water mass transported from the North Atlantic Ocean, and it has a relatively high concentration of $N_2O$ resulting from long-term accumulation via the deep-water nitrification process. However, significant supersaturation resulting from CDW upwelling is not observed at the surface water of Prydz Bay, whereas the north flank of the Antarctic Peninsula shows a higher $N_2O$ oversaturation since the upwelling of CDW at this site further approaches the surface layer because of the topography. These limited findings suggest that the upwelling of CDW during summer may not cause stratification and likely releases large amounts of $N_2O$ into the atmosphere.

The $N_2O$ concentration in the deeper part of the Southern Ocean ranges from 17.0–18.0 nmol·L$^{-1}$. Zhan et al. (2015b) suggested that the $N_2O$ concentration may result from the mixing process between CDW and Antarctic shelf water, which not only explains how this concentration can be observed at this depth but also suggests that the mixing process in the high-latitude Southern Ocean may provide a path for downward transportation of $N_2O$ and a mechanism for removing $N_2O$ from the atmosphere.

### 3.3 Implications of the $N_2O$ distribution in polar and sub-polar oceans

$N_2O$ is a relative inert chemical compound that accumulates along the path of thermohaline circulation; therefore, it shows unique variations across the oceans. The Greenland Sea (GS) and the Bering Sea (BS) are both Arctic-adjacent seas and sub-Arctic seas. As shown in Figure 2, the $N_2O$ concentrations in the GS (white triangles) are more vertically homogeneously distributed than those in the BS (gray triangles). The Aleutian Basin of the BS, indeed, is virtually part of the North Pacific Ocean, which is the oldest water in the global ocean. The $N_2O$ concentration is identical to that of the North Pacific Ocean (Butler et al.,

1989; Zhan et al., 2013). The highest $N_2O$ concentration in the BS is at ~900 m, which corresponds to the oxygen minimum zone, which is the oldest water in the global ocean. The $N_2O$ concentration in the GS is ~2 nmol·$L^{-1}$ higher than that in the CB, indicating the age difference between these two water masses. The oldest water in the GS is approximately 40 years old, whereas the deep water in the CB may be 300–500 years old (Zhan et al., 2015a). Zhan et al. (2015a) reveals that the $N_2O$ concentration in a certain water parcel and the age of this parcel are significantly correlated. A simulation of the $N_2O$ concentration in the intermediate CB water and deep CB water using a model based on the above correlation presented results that correspond well with the field observation results, thus showing that $N_2O$ can be used as a tracer for ocean circulation.

The $N_2O$ concentration difference between the CB and BS in the 3000 m to 4000 m interval is approximately 10 nmol·$L^{-1}$. Assuming that the accumulation rate is 0.0057 nmol·$L^{-1}$ (Bange and Andreae, 1999), the difference in the age between the BS deep water and CB deep water is approximately 1754 years, which is very close to that from the evaluation by Bange and Andreae (1999). However, Bange and Andreae (1999) mainly studied the water below 2000 m, and a deep water formation may occur in the Southern Ocean, even in the Aleutian Basin (Warner and Roden, 1995), which may bring relatively "new" $N_2O$ to the bottom water. Hence, it is important to estimate the contents of $N_2O$ in the deep ocean. The answer to this question will help to better constrain not only the production of $N_2O$ in the deep ocean but also the $N_2O$ sink strength of high-latitude oceans.

# 4 Source/sink characteristics of the polar and sub-polar oceans

The contribution of oceans to the global $N_2O$ budget is approximately 4 Tg N·$a^{-1}$; however, the role of polar and sub-polar oceans is not well constrained. The contribution of $N_2O$ from the Arctic Ocean has been previously neglected, which is probably because this ocean is mostly covered by sea ice. However, global warming has led to sea ice retreat, thus exposing more Arctic Ocean water, which may result in changes to the source/sink characteristics of the Arctic Ocean. Wu et al. (2017) showed that the BS did not show obvious source or sink characteristics while the continental shelves (both BS and Chukchi Sea) and open Arctic Ocean showed source and sink characteristics during the expedition (Table 1). However, the "sink" characteristic is probably a seasonal phenomenon caused by the seasonal melting of sea ice. Due to its role as one of the deep convection centers, the GS is the only permanent $N_2O$ sink that has been discovered thus far. For the past 40 years, the sink strength of this region has been approximately −5.0 ± 0.4 μmol·$m^{-2}$·$d^{-1}$ to −4.4 ± 0.4 μmol·$m^{-2}$·$d^{-1}$ (Zhan et al., 2016).

**Table 1** The air-sea fluxes of polar and sub-polar oceans

| | Sea area | Flux ($N_2O$)/(μmol·$m^{-2}$·$d^{-1}$) | References |
|---|---|---|---|
| Arctic and sub-Arctic | Chukchi Sea | 11.8 ± 9.1 | Hirota et al. (2009) |
| | Bering Sea | 2.1 ± 0.9 | |
| | Greenland Basin | −4.4 ± 0.4 | Zhan et al.（2016） |
| | Open sub-Arctic Ocean | 2.3 ± 2.2 | Zhan et al.（2020） |
| | Bering and Chukchi seas continental shelf | 3.7 ± 5.0 | |
| | Aleutian Basin | 0.46 ± 0.1 | Wu et al. (2017) |
| | Bering and Chukchi seas continental shelf | 8.2 ± 1.4 | |
| | Chukchi Abyssal Plain | −10.2 ± 1.4 | |
| Southern Ocean | Bellingshausen Sea and Drake Passage | −0.06 ± 0.9 – 0.09±1.4 | Rees et al. (1997) |
| | Antarctic Peninsula (sea area) | 8.33 ± 3.32 | Zhan et al.（2018） |
| | Prydz Bay | −0.3 ± 0.8 | Zhan et al.（2007） |
| | Prydz Bay (north of the shelf break） | −1.2 ± 0.44 | Zhan et al.（2015b） |
| | Prydz Bay (south of the shelf break) | −3.65 ± 0.95 | Zhan et al.（2015b） |
| | The high-latitude Southern Ocean | ＜9.8 ± 0.5 | Zhan et al. (2017) |

The Southern Ocean has been considered one of the most important oceanic $N_2O$ sources to the atmosphere as proposed by Bouwman et al. (1995), who assumed that the upwelling of $N_2O$-rich CDW in the Southern Ocean may be a significant source for atmospheric $N_2O$. Using historical data, Nevison et al. (1995) suggested that the Southern Ocean may contribute approximately 1/3 of the oceanic source; however, her study further revealed that the above evaluation may overestimate the source strength of the Southern Ocean. Thus, the contribution of the Southern Ocean has been approximated at 0.9 Tg N·$a^{-1}$ (Nevison et al., 2005).

To date, few air-sea-flux studies have been performed. The sea-to-air fluxes at the $N_2O$ undersaturation regions of Prydz Bay, the Drake Passage and the Bellingshausen Sea are −0.3 ± 0.8 μmol·m$^{-2}$·d$^{-1}$, ~3.7 ± 1.0 μmol·m$^{-2}$·d$^{-1}$ (Zhan and Chen, 2009; Zhan et al., 2015b) and −0.6 ± 0.4 μmol·m$^{-2}$·d$^{-1}$ – −0.9 ± 0.7 μmol·m$^{-2}$·d$^{-1}$ (Rees et al., 1997), respectively, suggesting weak sink characteristics; however, in the upwelling region in the Southern Ocean, a relatively strong source can be observed. The sea-to-air fluxes at the upwelling regions of the Bellingshausen Sea and the area north of the Antarctic Peninsula are 3.1 μmol·m$^{-2}$·d$^{-1}$ (Rees et al., 1997) and 8.8± 2.2 μmol·m$^{-2}$·d$^{-1}$ (Zhan et al., 2018), respectively. The $N_2O$ produced near the lower euphotic zone may also be a $N_2O$ source for the Southern Ocean (Law and Ling, 2001). Considering that the upwelling of CDW may occur in certain regions with a limited area, its contribution to the global budget may be limited, whereas $N_2O$ production in the low euphotic zone may be limited but has a wider distribution. Therefore, the importance of these two sources and their contributions may need to be evaluated.

# 5 Conclusions and future work directions

Although the Southern Ocean is considered a significant source of $N_2O$, polar oceans are infrequently studied. However, both polar oceans show that small areas of regional $N_2O$ undersaturation can act as a possible temporal or even permanent sink for $N_2O$, which may not challenge the above conclusion, although the contribution of these areas should be further constrained. The accumulation rate of $N_2O$ in deep water is very low and linearly corresponds to the age of the water mass, thus facilitating evaluations of the production of the deep ocean and indicating that $N_2O$ is a useful tracer.

According to the advances that have been made, the following questions still need to be addressed:

(1) How much $N_2O$ will be released into the atmosphere at the upwelling site? What is the difference between the upwelling outcrop region and the non-outcrop region? Does active production of $N_2O$ occur in the lower euphotic zone of the Southern Ocean? If so, what is most important contributor to the $N_2O$ budget?

(2) What are the $N_2O$ source/sink characteristics of the sea-ice margin region of both poles? Will the ice-melting process together with the deep-water formation process provide a $N_2O$ sink in the Southern Ocean?

(3) How much $N_2O$ is produced in the relatively "young" water masses, such as the Antarctic Bottom Water (AABW), in the deeper part of the ocean?

To address these questions, high-resolution surface water $N_2O$ data of the upwelling region should be obtained to illuminate the upwelling-introduced $N_2O$ source pattern and constrain the $N_2O$ source contributions. Moreover, powerful tools, such as isotope and molecular biological techniques, should be applied to reveal the complicated $N_2O$ production mechanism at the sea ice margin and below euphotic layer and more profile data that capture the $N_2O$ signal of certain water masses, such as CDW or AABW, should be acquired. With all these efforts, the $N_2O$ source/sink characteristics of the Southern Ocean and their role in $N_2O$ circulation will be further addressed.

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