

Marine biogenic aerosols and their effects on aerosol-cloud interactions over the Southern Ocean: a review

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Abstract The Southern Ocean (SO) plays an important role in the global climate system. Changes in SO biogeochemistry and marine ecosystems may influence the distribution of atmospheric aerosols and clouds and impact the climate system. We reviewed current knowledge on the interactions between marine aerosols and clouds over the SO. We focused on marine primary and secondary organic aerosols and summarized their characteristics, processes and roles as cloud condensation nuclei and ice nuclei. We described in detail the interactions between the marine ecosystem, aerosols and clouds. We discussed marine productivity, formation of marine biogenic aerosols and interactions between aerosols, clouds and climate. We explored the impact of climate change on SO marine ecosystem productivity and aerosol–cloud–climate feedback. Marine biogenic aerosols could impact the radiation budget and oceanic low-level clouds over the SO. This study contributes towards an improved understanding of marine productivity, aerosol-cloud interactions and climate change in the SO. The SO may respond to climate change in varying degrees. More studies are urgently needed to support accurate forecasts of future changes in the SO.

Keywords Southern Ocean, marine biogenic aerosols, cloud condensation nuclei, ice nuclei, climate change

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1 Introduction

Aerosols and clouds absorb and scatter solar and terrestrial radiation and play important roles in the Earth's energy balance. Cloudiness is closely related to the production, transport, and evolution of aerosols in the atmosphere. The Southern Ocean (SO) is one of the cloudiest regions on Earth; it is subject to limited continental and anthropogenic influence because of its great distance from the centers of industrial activities. As a result, the remote SO is an ideal location for the study of aerosol-cloud interactions. Clouds formed over the SO are highly sensitive to aerosol

perturbations (Koren et al., 2014). Understanding of aerosol formation over the SO and the impact of aerosols on cloud droplet concentrations is key to accurate simulation of cloud radiative effect, ocean temperature changes and other global climate parameters under future climate scenarios (Revell et al., 2019; Hamilton et al., 2020; McCoy et al., 2021).

Cloud droplets form on atmospheric aerosol particles; atmospheric aerosols are suspended solid and liquid particles with complex compositions and properties, which determine cloud properties (McCoy et al., 2015). There are two types of marine organic aerosols: primary organic aerosols (POAs) and secondary organic aerosols (SOAs); the production mechanisms and properties of both types are complex and well established. POAs are mainly composed of organic compounds from sea spray; their production

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mechanisms include bubble formation by breaking waves to remove surface-active organic matter and other substances (e.g. bacteria, viruses, and detritus) along the ascending trajectory, and then inject into the atmosphere as marine POAs at the time of the bursting of the bubbles (Blanchard and Woodcock, 1957; Blanchard, 1964; Barger and Garrett, 1970). SOAs are generally produced from the oxidation of volatile organic compounds (VOCs), such as dimethyl sulfide (DMS), emitted from the ocean. VOCs, such as monoterpenes, sesquiterpenes, oxygenated hydrocarbons, and isoprene, which accounts for nearly 50% of the total biogenic VOCs emissions, are the main hydrocarbon gases in the atmosphere (Guenther et al., 2006; Sindelarova et al., 2014). Origins of these organic aerosols include regional phytoplankton blooms, unstable organic matter in the surface ocean, and long-term global processes, such as upwelling of old refractory dissolved organic matter in the deep sea (Brooks and Thornton, 2018). Organic nitrates, such as methyl nitrate, emitted from the ocean could also be a source of organic aerosols (Ng et al., 2017); nitrate radicals (NO_3) may react with biogenic VOCs to form SOAs and organic nitrates (von Kuhlmann et al., 2003; Horowitz et al., 2007; Fry et al., 2014; Boyd et al., 2015). However, at present, several aspects concerning marine VOCs remain poorly understood; these include source intensity, dependence of VOCs on environmental variables such as temperature, salinity and insolation, and the mechanisms through which VOCs are chemically transformed into aerosols (Carpenter et al., 2012; Qu et al., 2018; Enami et al., 2019).

The SO is one of the cloudiest places on Earth because of the continuous passage of extratropical cyclones and their associated frontal systems (Stubenrauch et al., 2013). Low-level clouds are dominant over the SO (Alexander and Protat, 2018); they play a key role in regulating sea-air interactions and sea surface temperature changes (Takahashi and Hayasaka, 2020). Satellite and surface-based observations indicate that supercooled liquid water clouds and mixed-phase clouds are particularly common in the SO (Choi et al., 2014; Storelvmo et al., 2015; Ceppi et al., 2016); this may be the result of relatively low concentrations of ice nucleating particles (INPs) over the SO (Burrows et al., 2013; McCluskey et al., 2018). In the absence of INPs, production of primary ice in clouds may be suppressed, which limits precipitation (Alexander et al., 2021). Response of the low-level clouds over the SO remains an important source of uncertainty in climate simulations. The uncertainty mainly stems from the lack of consensus over cloud albedo changes from the different models (Zelinka et al., 2012a, 2012b, 2013).

The SO surrounds the Antarctic continent and connects the Pacific Ocean, the Atlantic Ocean, and the Indian Ocean. It comprises the Weddell Sea, Bellingshausen Sea, Amundsen Sea, Ross Sea, Pacific Ocean and the Indian Ocean; it accounts for 22% of the global ocean area and plays a unique role in global biogeochemical processes

(Chen and Lu, 2016; Chen et al., 2017). The SO is covered by seasonal sea ice, which alters the characteristics of the mixed layer. Figure 1 shows the maximum sea ice extent in the SO (Pellichero et al., 2017). On the basis of hydrographic data collected on research cruises, the frontal system of the SO is divided into (from south to north) the Subtropical Front, the Sub-Antarctic Front, the Polar Front, the Southern Antarctic Circumpolar Current Front, and the Southern Boundary Front. The fronts are defined by different water masses, which have important implications on the physical oceanography and the climate of the region (Chapman et al., 2020). On the basis of measurements of surface water temperature and salinity, the SO is divided into the Seasonal Ice Zone (SIZ) (south of 60°S), the Sub-Antarctic and Antarctic Zone (SAAZ) (60°S – 40°S), and the South Subtropical Zone (SSTZ) (40°S – 30°S) (Zhang et al., 2021); these divisions are shown in Figure 1.

The integrated system of atmosphere, sea ice, ocean and ecology of the SO plays a key role in regulating the global climate and affects global primary production and carbon exports (Chen et al., 2018). In the SO, temperature is increasing and salinity and pH are decreasing; the SO is the world's largest ocean carbon sink but its carbon storage capacity is declining (Ma et al., 2012). These changes will undoubtedly have profound impacts on global climate. Therefore, future research needs to focus on the dynamic interactions between the SO and the global climate system. Improved understanding of the connection, response, and feedback between atmosphere, ocean, ecosystem and climate change will contribute towards enhancing the next generation of global climate models.

Aerosols can act as cloud condensation nuclei (CCN) and ice nuclei (IN) and influence precipitation and cloud formation, water content, optical properties, and microphysical processes; they can even change the local and regional climate, especially over the remote SO. In this review, we summarize the latest research on marine biogenic aerosols and marine productivity in the SO, nucleation mechanisms in different ecosystems, aerosol-cloud interactions, and factors influencing these interactions. We discuss the impacts of climate change on marine ecosystem productivity and aerosol–cloud–climate feedback over the SO. Our aim is to provide a foundation for future studies of marine biogenic aerosols over remote oceans.

2 Marine ecosystem productivity in the SO

The SO is the largest high nutrient and low chlorophyll (HNLC) oceanic region on Earth; high concentrations of nitrate, phosphate, and silicate are maintained throughout the year (Watson, 2001). Nitrate concentration is high in surface waters along coasts and in the SO; seawater NO_3^- concentration increases poleward; in the SO, atmospheric

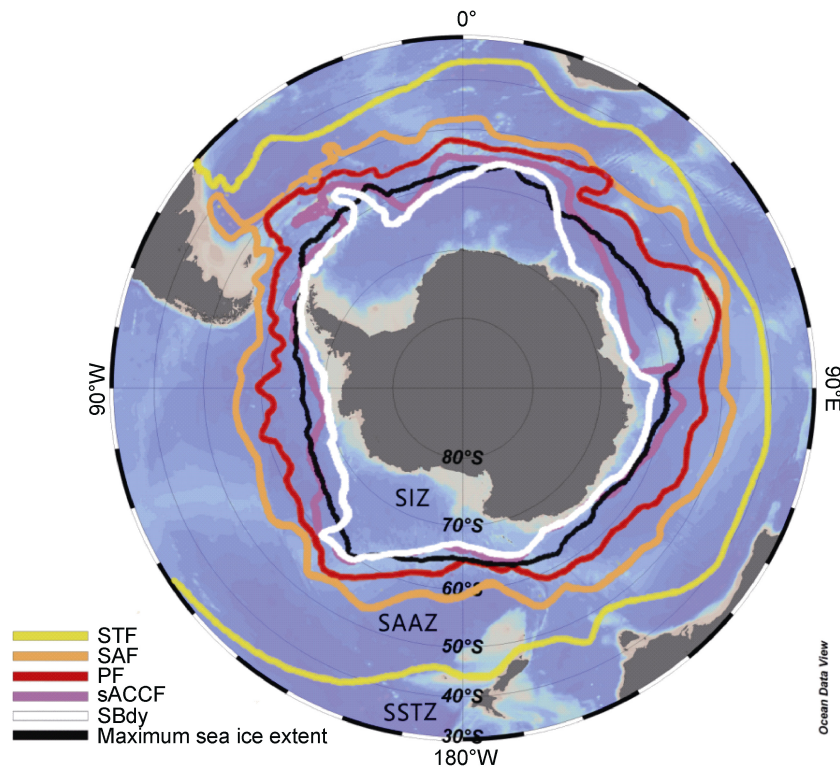


Figure 1 The Southern Ocean. The concept of the Antarctic Circumpolar Current and its fronts has evolved to include the Subtropical Front (STF), the Sub-Antarctic Front (SAF), the Polar Front (PF), the southern Antarctic Circumpolar Current Front (sACCF) and the Southern Boundary (SBdy) front. Solid black line indicates maximum sea ice extent. The Southern Ocean is divided into the following sections: south of 60°S, which is in the Seasonal Ice Zone (SIZ); 60°S–40°S, which is in the Sub-Antarctic and Antarctic Zone (SAAZ), and 40°S–30°S, which is in the South Subtropical Zone (SSTZ). Figure and terminology are based on Pellichero et al. (2017), Chapman et al. (2020), Zhang et al. (2021).

NO_3^- is a fairly small source of seawater NO_3^- (Shi et al., 2021). Using satellite data from the ocean surface layer, aerosol optical depth and cloud microphysical parameters, Meskhidze and Nenes (2010) found significant positive correlations between marine ecosystem productivity, submicron aerosol abundance, and cloud microphysical properties in different oceanic regions. Seawater mixing and advection, solar radiation and seasonal sea ice changes induce changes in light, seawater temperature and phytoplankton uptake rate of nutrients (e.g., nitrate and silicate). Ardyna et al. (2017) reported that these changes alter phytoplankton biomass and phenology and directly determine the primary productivity of the SO.

Marine productivity is under the influence of several factors. Iron is generally considered to be a limiting factor of phytoplankton productivity in HNLC regions (Tagliabue et al., 2017); its limiting effect on phytoplankton in the Ross Sea has been confirmed by enrichment experiments (Sun et al., 2009). Duprat et al. (2016) reported that the melting of an iceberg resulted in the input of terrigenous nutrients and trace elements into the surrounding waters, which increased the primary productivity of the area within several kilometers of the iceberg. Diatoms are one of the main producers in the SO ecosystem. The zinc silicon record of

diatom opal in Prydz Bay indicates that marine productivity has increased over the last century; future temperature increase, sea ice decline or increase in iron supply could lead to significant increase in diatom productivity (Sun et al., 2016). O'Dowd et al. (2015) reported that phytoplankton biomass, chlorophyll-*a* and net primary productivity determine the mass fraction of organic matter enriched in sea spray; these results suggest that there is an important coupling between biologically-driven plankton bloom termination, marine productivity and sea spray modification, and have potential implications for climate change. Behrenfeld et al. (2006) found considerable variations in ocean productivity between the 1990s and the 2000s, which have been most likely caused by climate-induced changes in marine stratigraphy and nutrient supply. Results from laboratory experiments suggest that ocean warming may increase primary production and efficiency in the SO (Feng et al., 2010; Zhu et al., 2016). In contrast, ocean acidification may reduce productivity by changing the chemical environment of the plankton community (Orr et al., 2005; Shi et al., 2010). Future increases in light, iron and temperature are likely to result in increased primary production at high altitudes (Hauck et al., 2015; Leung et al., 2015; Moore et al., 2018). Marine

primary production underpins the carbon cycle, air-sea carbon dioxide (CO₂) exchange and ecosystem functions, which is important for the operation of the global biogeochemical system, especially in the context of climate change (Falkowski and Raven, 2007).

3 POAs and SOAs over the SO

The SO is the most pristine region on Earth for the measurement of aerosols (Hamilton et al., 2014). In the pristine marine environment, aerosols are the results of primary and secondary processes. Aerosols over remote oceans are mixtures of sea salt, sulfate and organic matter, which are from anthropogenic and natural sources (e.g., dust, sea salt, soot, and biological particles) or are products of VOCs oxidation (Andreae, 2007; Andreae and Rosenfeld, 2008). Analyses of NO₃⁻ isotopes indicate that

elevated atmospheric nitrate concentrations in coastal areas are related to human activities, while nitrate concentrations in high southern latitudes are related to precursor Antarctic snowpack emissions driven by photolysis (Shi et al., 2021). Table 1 shows the properties of marine biogenic aerosols and gas precursors over the SO. Rinaldi et al. (2020) measured aerosol composition near the Antarctic Peninsula and reported that water-soluble organic matter, excluding methanesulfonic acid (MSA), accounts for 6%–8% and 11%–22% of the PM₁ mass in the open ocean (OO) and sea ice (SI) areas, respectively; other components include sea salt (86%–88% OO, 24%–27% SI), non-sea-salt sulfate (nss-SO₄²⁻) (3%–4% OO, 35%–40% SI) and MSA (1%–2% OO, 11%–12% SI). Studies of natural aerosols in pristine environments will help us to further understand how aerosols affect the climate and the Earth system (Hamilton, 2015).

Table 1 Marine aerosol and gas precursor properties and microphysical parameters related to aerosol-cloud interactions over the Southern Ocean

Property	Value	Reference
DMS in marine atmosphere/(ng·m ⁻³)	899.8±957.9	
Particulate MSA/(ng·cm ⁻³)	30.6±16.8	Yan et al., 2020
SO ₄ ²⁻ in aerosol phase/(ng·cm ⁻³)	148.1±32.5	
Biogenic SO ₄ ²⁻ /(ng·m ⁻³)	47.1±30.2 (up to 163.8)	
Particulate MSA/(μg·cm ⁻³)	0.13 (0.08–0.23)	
MSA gaseous/(molecules·cm ⁻³)	2.6×10 ⁶ (1.0×10 ⁶ –6.2×10 ⁶)	Schmale et al., 2019
CCN _{0.2} /cm ⁻³	105 (65–150)	
DMS concentration of seawater/nM	3–12	
DMS flux of seawater/(μmol·m ⁻² ·d ⁻¹)	27 (1–101)	Gabric et al., 2018
Biogenic SOAs tracers/(pg·m ⁻³)	106±68.5 (2.26–303)	Deng et al., 2021
Biogenic contribution to CCN	35% (winter) – 80% (summer)	Vallina et al., 2006
CCN column content/cm ⁻²	2.0×10 ⁸ to more than 5.0×10 ⁸ for a chlorophyll increase from 0.3 to about 0.5 mg·m ⁻³	Krüger and Graßl, 2011
Aerosol number concentration/cm ⁻³	50–100 (winter) to 700 (summer maxima)	Bromwich et al., 2012

Sea spray POAs consist of biogenic POAs and gel-forming organic materials, which are primarily carbohydrates and related semi-volatiles (Brown et al., 1964; Bauer et al., 2003; Facchini et al., 2008; Hawkins and Russell, 2010). Biogenic POAs include fungal spores, bacteria, viruses, microbial fragments, pollens and plant debris, which are ubiquitous particles that are emitted directly from the biosphere into the atmosphere (Perrino and Marcovecchio, 2016; Amato et al., 2017; Zhai et al., 2018; Hu et al., 2020). In the atmosphere, microorganisms are ubiquitous and highly diverse. Globally, most aerosols are produced by bubbles bursting at the air-sea interface; the particles produced from the bursting of bubbles account for a large proportion of POAs in terms of particle numbers. Monahan et al. (1986) found that sea spray is primarily generated by the air-sea exchange associated with the breaking of waves and bubbles at the ocean surface. Sea

spray droplets evaporate in the air and gather in the sea surface microlayer. The remaining aerosol particles are mainly composed of sea salt and organic particles (Quinn et al., 2014). Uetake et al. (2020) found that bacteria in the SO boundary layer mainly come from sea spray and contain very low contributions from Australian and Antarctic soils. Myriokefalitakis et al. (2010) found that marine POAs are the main contributors to submicron organic aerosol mass. In terms of mass concentration, sea spray aerosol (SSA) produced by wind-driven process is one of the largest sources of primary atmospheric aerosol particles (Warneck, 1988). By analyzing the size distributions of POAs, Gantt and Meskhidze (2013) found that sea salt is absent from fine marine POAs (diameters below 200 nm) and is mixed into coarse POAs (diameters above 1000 nm). Chlorophyll-*a* concentration is the most commonly used indicator of plankton biomass. Lana et al. (2012) suggested

proportionality between POAs concentration and chlorophyll-*a* concentration, and between POAs concentration and plankton biomass. While Mayer et al. (2020) believed that only secondary marine aerosols are correlated with phytoplankton biomass. Ovadnevaite et al. (2011) found that marine POAs have a low hygroscopic growth factor (~ 1.25) but an activation efficiency that is higher than that of inorganic aerosols that are more soluble. However, to date, there have been few measurements of POAs over the SO, and more studies need to be conducted in the future.

There are many potential precursors of marine SOAs; these include isopentene, terpenes, amines, alkylnitrates, and alkanes (Bonsang et al., 1992; Chuck et al., 2002; Yassaa et al., 2008). Using a parametric model and forecast data, Myriokefalitakis et al. (2010) computed the fraction of global marine SOAs from DMS oxidation ($\sim 78\%$), possible dialkyl amine salt ($\sim 21\%$), and marine hydrocarbon oxidation ($\sim 0.1\%$). MSA and nss-SO_4^{2-} produced by the oxidation of DMS considerably impact the CCN in the marine boundary layer and are of great significance to the biological sulfur cycle and climate change (Yan et al., 2020). The microphysical parameters related to DMS and oxidation products are shown in Table 1. Studies on marine SOAs have mainly focused on MSA formed by the oxidation of DMS; limited attention has been paid to the contribution of isoprene and monoterpenes to SOAs (Arnold et al., 2009; Gantt et al., 2009; Luo and Yu, 2010; Myriokefalitakis et al., 2010). Hu et al. (2013) pointed out that marine isoprene emissions can abruptly increase during phytoplankton blooms; as a result, isoprene is an important source of organic aerosol over the ocean. The SOAs produced by isoprene are considered to be the single largest source of atmospheric organic aerosols (Fang et al., 2012).

Figure 2 shows the process of SOAs formation from isoprene mediated by cloud processes; the process may include the distribution of isoprene oxygenated products such as glyoxal, methylglyoxal and pyruvic acid in cloud water, and the formation of glycolic, glyoxylic and oxalic acid via oxidation (Myriokefalitakis et al., 2010). Studies of these important SOAs precursors will help improve our understanding of physical and chemical processes in the atmosphere (Fang et al., 2012). Over the SO, there are diverse species of SOAs. Concentrations of MSA along the coast of East Antarctica are higher than those over the SO, possibly because DMS emissions from marine biological sources along the Antarctic coast are higher than those over the SO (Xu et al., 2013). In the nutrient-rich upwelling regions along the Antarctic coast and the SO convergence, high biological activity increases DMS flux; this may lead to increases in the production of nss-SO_4^{2-} or MSA aerosols (Simpson et al., 2014; Alroe et al., 2020). Between austral spring and summer, seawater DMS and dimethylsulfoniopropionate concentrations generally increase in the two zones of the SO (SIZ and SAAZ in Figure 1) (Curran and Jones, 2000). In terms of atmospheric MSA, concentration can also be influenced by production mechanism. Antony et al. (2010) found that MSA concentrations in ice-covered regions are significantly higher than those in ice-free regions along the coast of Larsemann Hills, East Antarctica. Nutrients in the snow stimulate algae growth; compounds produced during algae growth lead to photoactivated reactions on aerosols and the snow surface, which promote the production of oxidants and increase MSA concentrations (Read et al., 2008; Antony et al., 2010). Curran et al. (2003) reported that high MSA concentrations are associated with large sea ice extents. The detailed mechanism of this relationship is

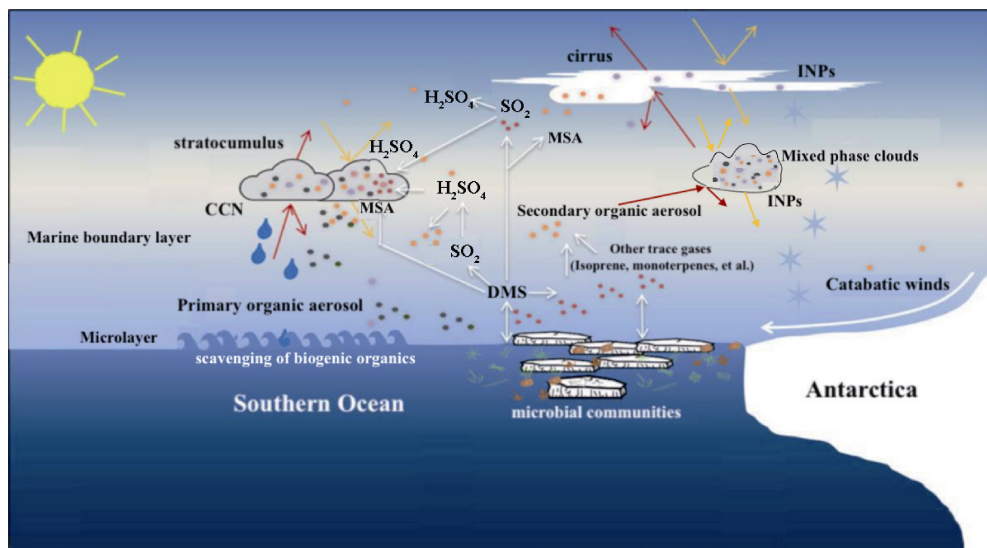


Figure 2 Processes of biogenic primary and secondary organic aerosols over the Southern Ocean in austral summer (modified from Schmale et al. (2019)).

complex. Xu et al. (2013) reported that most MSA was enriched in the accumulation mode, and only a small fraction was enriched in the coarse mode. Different physical and environmental factors such as variability in solar radiation intensity, precipitation patterns, sea ice conditions, winds and ocean currents may affect MSA properties. Xu et al. (2013) also found that oxalate was mainly accumulated in the fine mode over the SO and East Antarctica; they found acetate in only a few samples from the coastal regions of East Antarctica; they reported that succinate over the SO has a bimodal size distribution. Mayer et al. (2020) found a strong positive correlation between phytoplankton biomass (i.e. chlorophyll-*a* concentrations) and secondary marine aerosols composed of sulfate, ammonium, and organic species. Phytoplankton functional groups may affect overall isoprene production, influencing the concentration of marine aerosols (Sanchez et al., 2021b).

4 Aerosol-cloud interactions over the SO

Researchers in atmospheric science are interested in understanding the interactions of aerosols with cloud and precipitation, because these interactions impact the water cycle, the atmosphere, as well as the climate system. Aerosol particles participate in the Earth's radiation budget and the climate system via two pathways: radiative forcing due to aerosol-radiation interactions (RFari) that arise from direct scattering and absorption of solar and terrestrial radiation by aerosols and radiative forcing from aerosol-cloud interactions (RFaci) that arise from the role aerosols play as CCN and IN; RFari and RFaci have partly offset the warming caused by greenhouse gases since the pre-industrial era (Yang et al., 2011; Lohmann, 2015). As CCN or atmospheric IN, aerosol particles could affect cloud microphysical processes, which then influence rain, snow, hail, and other forms of precipitation. Research and development of weather modification and climate prediction techniques are supported by improved understanding of the influence of aerosols on cloud macro- and micro-structure and precipitation, and improvements in cloud detection (Li et al., 2014). Mixed-phase stratocumulus with supercooled droplets, including cloud liquid droplets and ice crystals, lies above the SO (Figure 2) (Huang et al. 2015). Ice crystals form on INPs at temperatures higher than the homogeneous freezing point of water (at <-36 °C), while liquid droplets form on CCN (Schmale et al., 2019). In Sections 4.1 and 4.2, we summarize recent research progress on aerosol-cloud interactions over the SO, and discuss the role of marine biogenic aerosols as CCN or IN over the SO.

4.1 Aerosols as CCN over the SO

Schmale et al. (2019) reported that biogenic emissions play an important role in CCN abundance in the Ross and

Amundsen seas. During phytoplankton blooms, microbial activity is associated with high CCN number concentrations. Table 1 shows that marine ecosystems (especially those in biologically active areas) can have considerable effects on CCN number concentrations over remote oceans (Meskhidze and Nenes, 2010); CCN number concentrations over remote oceans typically range from a few tens per m^3 in biologically inactive regions to a few hundred per m^3 in biologically active regions (Andreae, 2007). Biogenic contributions to CCN are estimated to be between 35% (winter) and 80% (summer) (Vallina et al., 2006). Lana et al. (2012) showed that, northward of 30° N and southward of 30° S, CCN seasonality matches with the seasonality of the production flux of marine secondary aerosols (DMS-derived and SOAs) but not with that of primary aerosols (POAs and sea salt), except over a narrow band over the SO. Without anthropogenic influence, oxidation products from marine-emitting DMS, i.e., $nss-SO_4^{2-}$ and MSA, can be responsible for very large proportions of the CCN population (Schmale et al., 2019). These findings confirm the central role of biogenic DMS emissions in controlling the quantity and variability of CCN over remote oceans. For sulfur and SOAs, positive correlations with CCN numbers and negative correlations with cloud droplet effective radius (r_e) are very common at the middle and high latitudes; these results indicate the importance of sulfur and SOAs in seeding cloud droplet activation; in contrast, primary aerosols (organic and sea salt) are positively correlated with CCN only at low latitudes; the correlation between primary aerosols and r_e varies, which indicates that primary aerosols are not the main driver of cloud microphysical changes, although they are large contributors to marine aerosol mass (Lana et al., 2012). Data collected on research cruises in the SO indicate that the contribution of SSA to CCN can vary between 10% and 100% (Quinn et al., 2017; Fossum et al., 2018). The coarse mode sea salt generated by wind is an important CCN component; between autumn and mid-spring, it contributes to approximately 36% of the CCN population (Gras and Keywood, 2017). Vallina et al. (2006) reported that CCN concentrations are high in austral summer and low in austral winter; on the basis of multiple linear regression models, they concluded that the seasonal and interannual variabilities of potential CCN are related to chlorophyll, the hydroxyl radical, and rainfall changes, and are independent of sea salt particles because chlorophyll could be regarded as the emission index of the planktonic aerosol precursor DMS and the hydroxyl radical could be considered as the emission index of the oxidant of DMS.

Marine POAs and sea salt can be mixed externally or internally. The effect of external mixtures of marine POAs and sea salt on CCN and cloud droplet number concentration (CDNC) is much greater than the effect of internal mixtures on CCN and CDNC; emissions of externally-mixed marine POAs result in a 1.3% increase in global CCN surface concentrations (Meskhidze et al., 2011;

Gantt et al., 2012). Over biologically productive parts of the ocean, introduction of externally mixed marine organic aerosols increases the yearly-average CCN concentrations by up to 20%, while introduction of internally-mixed aerosols reduces CCN concentrations (Meskhidze et al., 2011). Increasing the water-soluble organic carbon mass in different sea salt and organic matter mixture models, O'Dowd et al. (2004) reported that CDNC increased by about 15%–20% in the internal mixture model and by more than 100% in the external mixture model.

Cloud condensation nuclei number concentration is also affected by aerosol hygroscopicity. Petters and Kreidenweis (2007) suggested that the influence of aerosol hygroscopicity on CCN activity can be measured using the hygroscopicity parameter, κ ; κ is 0.5–1.4 for highly-CCN-active salts, 0.01–0.5 for slightly to very hygroscopic organic matter, and around 0 for non-hygroscopic components. Other studies reported that high hydrophobicity is associated with low CCN activity; these findings have resulted in minor changes in CCN number concentrations and cloud properties associated with marine organic aerosols (Fuentes et al., 2010; Moore et al., 2011; King et al., 2012). For internally-mixed sea salt aerosols, replacing highly hygroscopic sea salt by less hygroscopic organic matter reduces surface CCN concentrations by 5%; marine organic compounds could induce surface tension depression, which reduces CCN number concentrations (Moore et al., 2008; Westervelt et al., 2012).

Furthermore, CCN concentration is independent of wind speed except in the most pristine regions, such as the SO. Vallina et al. (2006) reported little change in wind speed over the SO except for slight increases winter, which is consistent with seasonal sea salt changes. Dunne et al. (2014) found that the impact of wind speed changes on marine CCN may be insignificant except in the most pristine regions. In most marine regions, CCN is mainly controlled by the effects of wet scavenging and continental pollution, and the effects of wind speed on CCN are negligible. Only in the most pristine regions, changes of wind-speed-driven marine aerosol emissions may generate climate feedback via clouds. Because the SO is far from continental sources of pollution, CCN can be expected to come primarily from natural sources, and the impact of wind speed trends on CCN concentration is more likely to be detected. Moreover, feedback via changing precipitation pattern and intensity may occur over most oceanic regions, because nucleation scavenging has the largest absolute effect on CCN concentration (Dunne et al., 2014). Sanchez et al. (2021a) found that, over a 1.5-day trajectory, precipitation is inversely related to CCN concentration, which indicates that precipitation scavenging is one of the main CCN sinks in the SO.

Soluble compounds and the atmospheric processes involved in their formation may be a source of CCN in the atmosphere. Some studies proposed that organic acid could

contribute to CCN formation under certain conditions (Pradeep Kumar et al., 2003; Abbatt et al., 2005). Organic acid could be produced directly from the oxidation of biogenic VOCs. For example, in gas phase, the photochemical decomposition of isoprene produces formic acid, methacrylic acid and pyruvic acid (Jacob and Wofsy, 1988). Although 98%–99% of these volatile organic acids are found in the gas phase, their concentrations in aerosol particles are sufficient to make them good candidates for CCN. Organic acids may contribute to the indirect (cloud-mediated) forcing caused by aerosols (Yu, 2000).

In austral summer, biogenic emissions from Antarctic coastal areas could explain the bulk of CCN and CDNC, while primary marine aerosols account for only about 20% of observed CCN and CDNC; owing to the remoteness of the SO, coastal biogenic emissions appear to be the main CCN source of the marine boundary layer, which suggests that, under climate change, variations in phytoplankton production along the Antarctic coast may have significant impacts on CCN concentrations and cloud characteristics over the SO (Sanchez et al., 2021a).

4.2 Impacts of marine biogenic aerosols on IN

Oceanic low-level clouds are mostly supercooled liquid water and considered as ice in many climate models (e.g. Bodas-Salcedo et al., 2012). The process of maintaining supercooled liquid clouds over the SO is complex and not well constrained (Gettelman et al., 2020); mixed-phase clouds are ubiquitous and play a significant role in the global radiation budget, especially at high latitudes (Shupe et al., 2008). Aerosol particles can act as CCN or IN in the formation of mixed-phase clouds; INPs can cause mixed-phase clouds to glaciate and alter their physical properties (Lohmann, 2002; Lohmann and Feichter, 2005; DeMott et al., 2010). Tan et al. (2016) reported that low clouds over the SO are sensitive to ice nucleation and vapor deposition (the Wegener–Bergeron–Findeisen or WBF process). Vergara-Temprado et al. (2018) found that the stratocumulus in the cold zone of the SO is sensitive to the ice nucleation scheme.

Ice nucleating particles trigger the freezing of supercooled cloud droplets, which leads to changes in cloud reflectivity, the amount of energy in the different layers of the atmosphere, and the amount of radiation reaching the surface (DeMott et al., 2010). Studies of atmospheric INPs sources will help us to further understand the effects of atmospheric INPs on cloud lifetime, cloud albedo and precipitation (Hoose and Möhler, 2012; Murray et al., 2012). The majority of the Earth's surface is covered by oceans. Oceans are the source of SSA particles, which contain large amounts of organic materials. Therefore, SSA particles are significant contributors to the global radiation budget; SSA particles act as INPs; they induce freezing at temperatures that are higher than typical freezing temperatures, and hence, affect the structure and properties of mixed-phase clouds (Mitts et al., 2021). Mitts et al. (2021) suggested that

supermicron SSA particles are more efficient INPs than submicron SSA particles because supermicron particles could carry a greater number of ice active components; there are clear differences between aged and nascent SSA, and the distinction is likely to have a strong impact on oceanic clouds and climate. Wilson et al. (2015) proposed that large amounts of organic materials in SSA could nucleate ice under conditions that support the formation of mixed-phase and high-altitude ice clouds (Figure 2). Model simulations and measurements indicate the domination of marine organic aerosols in INPs concentrations in remote locations like the SO (Burrows et al., 2013; Wilson et al., 2015; Vergara-Temprado et al., 2017). Marine phytoplankton, such as *Prochlorococcus*, *Synechococcus*, and pico- and nano-eukaryotes, are also effective sources of deposition- and immersion-mode INPs (Ladino et al., 2016; Wolf et al., 2019; Wilbourn et al., 2020). Concerning atmospheric processes, certain microorganisms or their by-products, could also act as INPs (Schnell and Vali, 1976; Vali et al., 1976). Changes in biogenic IN concentrations may affect precipitation and cloud cover in remote marine areas and alter the Earth's hydrological cycle and energy balance (Burrows et al., 2013). Hartmann et al. (2020) reported that local marine sources of biogenic INPs at 80°N could effectively nucleate ice at relatively high temperatures even in winter. Schmale et al. (2019) found that IN concentrations in Antarctica are lower than those in marine air masses in the Northern Hemisphere; in Antarctica, IN concentrations decrease between austral summer and autumn, and the difference between open ocean and coastal samples is small. The number of particles that can act as IN increases sharply as temperature decreases; only a small fraction of aerosols could function as IN at all temperatures and considerable seasonal and spatial variability exists (Meyers et al., 1992; Murray et al., 2012). Our current understanding of ice nucleation is limited. Quantitative experiments are needed to assess the ice-nucleating ability of different materials (Murray et al., 2012). Future field studies of marine biogenic INPs in the SO are also needed to improve our understanding of the effects of SO marine biogenic INPs on the climate system.

5 Impacts of climate change on SO marine ecosystem productivity and aerosol–cloud–climate feedback

The polar regions are among the places that are changing the most rapidly on Earth as a result of global climate change (Steig et al., 2009; Hoegh-Guldberg et al., 2018). Climate change is expected to lead to major changes with significant impacts on nutrient supply and carbon circulation in the SO; many of these changes are likely to be already in progress (Hirst, 1999; Plattner et al., 2001; Landschützer et al., 2015; Swart et al., 2018). Under continuous increase in anthropogenic CO₂ emissions and global warming, ocean acidification, particularly in the

polar regions, is becoming an increasingly urgent issue. The polar oceans are large sinks of CO₂ and have fragile marine ecosystems; they will be significantly impacted under future ocean acidification (Yamamoto-Kawai et al., 2009; Beaufort et al., 2011). The SO has a large CO₂ absorption capacity; therefore, ocean acidification is more prominent in the SO than in other oceans. In the SO, there are significant spatial and temporal variations of the aragonite saturation state—an index used to estimate ocean acidification. Aragonite saturation state is high in summer and low in winter; it decreases with depth; it is lower in offshore areas and higher in the open ocean; these results indicate high levels of acidification in offshore surface waters (Wang et al., 2016).

In addition, there is a positive climate feedback mechanism, involving a decrease in CCN concentrations due to regional climate warming. Using a physics-based nucleation mechanism, Yu et al. (2012) showed that increase in global temperature could significantly inhibit the rate of formation of new particles and CCN. Under global warming, CCN decrease could lead to increase in the average size of cloud droplets, which would lead to the reduction of cloud albedo and cloud cover and the increase of precipitation (Twomey, 1977; IPCC, 2007). Decrease of cloud albedo and cloud cover increases the amount of solar radiation reaching the surface and creates a positive feedback mechanism that amplifies the initial warming. However, further verification is needed to evaluate the importance of this mechanism, because this analysis only considered the effect of temperature rise on the formation of new particles but ignored the effects of photochemical reaction rate, oxidant concentration and other related factors (Yu et al., 2012).

Climate-induced changes in Antarctic sea ice have received widespread attention. In terms of sea ice growth, Zhang (2007) proposed a negative feedback mechanism to explain the increase of Antarctic sea ice under global warming. This strong negative feedback counteracts the atmospheric warming in the Southern Hemisphere. Bintanja et al. (2013) suggested that accelerated basal melting of ice shelves probably contributes significantly to the increase of Antarctic sea ice. In the Northern Hemisphere, there has been a significant decrease in Arctic sea ice; in the Southern Hemisphere, there have been large interannual variations in Antarctic sea ice (Alley et al., 2007). Satellite data indicate that Antarctic sea ice extent has increased since the late 1970s; most of the increase has been in the western Ross Sea; there has been some increase in the Weddell Sea and the Indian Ocean; there have been sharp declines in the Bellingshausen and Amundsen seas (Turner et al., 2009; Sen Gupta et al., 2009). In contrast, Chen and Lu (2016) reported that, between 2009 and 2013, the largest increase in sea ice extent was found in the Indian Ocean sector of the SO. Many phytoplankton blooms occur in the open waters and the SIZ of the SO (Moreau et al., 2020) probably because of the inflow of nutrient-laden freshwater derived

from melting sea ice (Sabu et al., 2014). Climate change and changes in coastal water conditions may affect the metabolic capacity of local microorganisms and trigger dynamic changes in community composition (Montes-Hugo et al., 2008, 2009). Sea ice cover changes therefore have implications for ecosystems and climate at the global and regional scales (Hobbs et al., 2016). The melting of icebergs could reduce convective overturning and limit heat transport from the deep ocean to the surface, thereby accelerating sea ice production over much of the SO in autumn and winter (Marsland and Wolff, 2001). In the Bellingshausen Sea, sea ice thickness is decreasing possibly because of the advection of warmer waters flowing along the Antarctic Coastal Current (Merino et al., 2016). Decrease in sea ice thickness may interact with climate change and may (at least partially) be caused by large-scale changes in atmospheric circulation; this should be a key focus of future research (Vaughan et al., 2003; Ma et al., 2012; Parkinson and Cavalieri, 2012).

Future climate change may have considerable impacts on aerosols. The response of natural aerosols may amplify or attenuate the effects of forcing (Carslaw et al., 2010). Marine gas components of vegetation emissions can lead to the formation of SOAs in the atmosphere and are likely to vary under climate change. Observations and model simulations of DMS, aerosol sulfate, CCN and cloud properties indicate correlations between phytoplankton dynamics, aerosols and cloud microphysical processes (Ayers and Gras, 1991; Boers et al., 1994; Ayers et al., 1997; Korhonen et al., 2008). The effect of DMS on the climate systems mainly depends on the ability of the oxidation products of DMS to increase CCN and cloud drop numbers. Two possible changes in ocean DMS emission have been proposed. First, under atmospheric warming, the depth of the mixed layer is reduced; as a result, phytoplankton resides near the surface; the increased solar radiation and DMS emissions lead to a negative climate feedback (Vallina et al., 2007; Vallina and Simó, 2007a, 2007b). In contrast, Behrenfeld et al. (2006) proposed that warming-induced ocean stratification can reduce the supply of nutrients from deeper waters; plankton growth is limited, and results in a positive feedback.

In 1987, Charlson et al. put forward the CLAW hypothesis (named after the authors of the paper; Charlson et al., 1987), which is a possible negative feedback mechanism between phytoplankton and global climate change; DMS is emitted from the ocean into the atmosphere and generates sulfate aerosols through atmospheric nucleation; the aerosols could act as CCN and impact the atmospheric radiation budget (Figure 2). Using perturbation experiments, Gunson et al. (2006) calculated the sensitivity of global DMS flux to perturbations of sea surface temperature, wind speed and ocean DMS concentration; comparing the sum of these sensitivities with results from simulated perturbation experiments, they reported a feedback factor of 0.06, which indicates a small negative

feedback and is in support of the CLAW hypothesis. However, more recent research suggests that the DMS biological control on CCN may not exist, and the response of clouds to aerosol changes may be much more complex than that understood decades ago; CCN sources in the remote marine boundary layer are also far more complex than the CLAW hypothesis admitted (Quinn and Bates, 2011). Studies in the Arctic also questioned the key role of DMS in the CLAW hypothesis (Leck and Bigg, 2007). However, this does not rule out the possible link between marine microbes and climate. Instead, the links between marine biology, cloud properties, and climate may be more subtle and complex. In addition, many other marine biogenic emissions, such as emissions of organohalogens (Carpenter et al., 2003), ammonia (Schlesinger and Hartley, 1992), isoprene (Bonsang et al., 1992; Palmer and Shaw, 2005; Arnold et al., 2009), monoterpenes (Yassaa et al., 2008), and other non-methane hydrocarbons (Guenther et al., 1995) may also affect atmospheric chemistry and climate. Their effects on climate remain to be quantified, and their effects on oxidants and aerosol processes over remote marine areas should be assessed.

In the future, the SO will respond to the combined processes of ocean acidification, variations in sea ice, radiative forcing due to interactions of aerosols and clouds, and other mechanisms to different degrees. More observations and studies are in urgent need to support accurate forecasts of future changes in the SO.

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