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Multiple eco-regions contribute to the seasonal cycle of Antarctic aerosol size distributions

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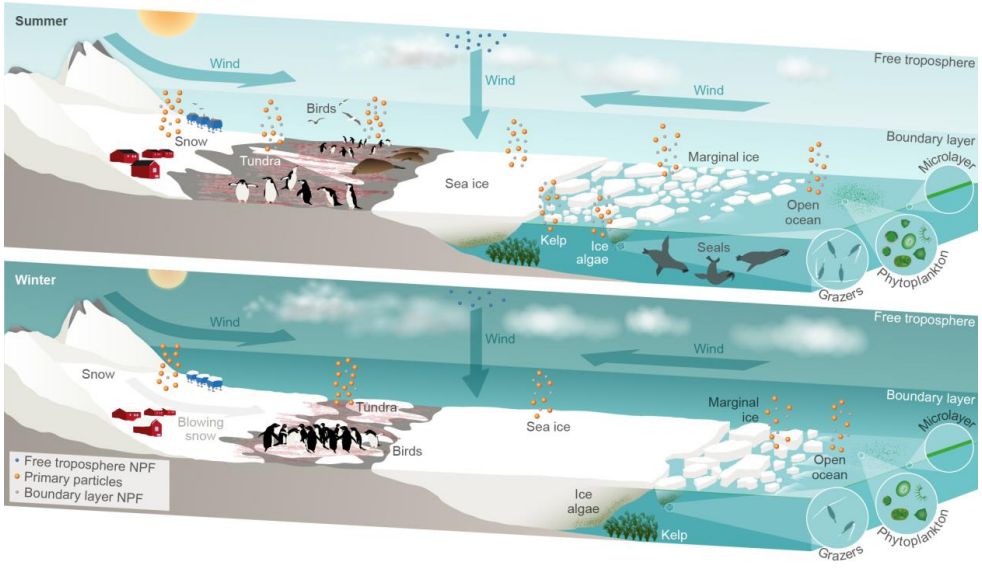
Graphical abstract



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Abstract

In order to reduce the uncertainty of aerosol radiative forcing in global climate models, we need to better understand natural aerosol sources which are important to constrain the current and pre-industrial climate. Here, we analyze Particle Number Size Distributions (PNSD) collected during a year (2015) across four coastal and inland Antarctic research bases (Halley, Marambio, Concordia/Dome C and King Sejong).

We find that the four Antarctic locations have striking differences in PNSD, stressing multiple aerosol sources and processes likely exist. We utilise k-means cluster analysis to separate the PNSD data into six main categories. Nucleation and Bursting PNSDs occur 28-48% of the time between sites, most commonly at coastal sites Marambio and King Sejong where air masses mostly come from the west and travel over extensive regions of sea ice, marginal ice, and open ocean, and likely arise from new particle formation. Aitken high, Aitken low, and bimodal PNSDs occur 37-68% of the time, most commonly at Concordia/Dome C on the Antarctic Plateau, and likely arise from atmospheric transport and aging from aerosol originating likely in both coastal boundary layer and free troposphere. Pristine PNSDs with low aerosol concentrations occur 12-45% of the time, most common at Halley located at low altitudes and far from the coastal melting ice, and influenced by air masses from the west.

The Antarctic Atmospheric circulation has a strong control on the air mass origin type. Most of the time Marambio and King Sejong stations are affected by Easterly air masses, whereas Halley gets air masses mainly from the Weddell sea marginal and consolidated pack ice. Not only the sea spray primary aerosols and gas to particle secondary aerosols sources, but also the different air masses impacting the research stations should be kept in mind when deliberating upon different aerosol precursors sources across research stations.

We provide evidence that both primary and secondary components from pelagic and sympagic regions strongly contribute to the annual seasonal cycle of Antarctic aerosols which add insight on the possible sources of aerosol production/activity in

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the whole Antarctic region. Our simultaneous aerosol measurements stress the importance of the variation in atmospheric biogeochemistry across the Antarctic region.

Introduction

The pristine region of the Southern Ocean (SO) plays a major role in modulating Earth's climate (Carslaw et al., 2013), and natural aerosols are one of the greatest sources of uncertainty in estimates of global radiative forcing (Reddington et al., 2017). This uncertainty becomes greater in polar regions (Carslaw et al., 2013). Aerosols modulate the climate both directly, by absorbing and reflecting radiation, and indirectly, by acting as cloud condensation nuclei (CCN), modulating cloud properties. The surface of the SO near the Antarctic continent goes through an annual freezing cycle; this large frozen area harbours one of the globes largest ecosystems providing a stable habitat for diverse microbial assemblages (Arrigo et al., 2009; 2015). Understanding these processes is key in polar environments where warming processes are rapidly affecting delicate ecosystems (Clem et al., 2020). For example, the Antarctic Peninsula has shown some of the largest increases in near-surface air temperature measured globally across the last 50 years (Turner et al., 2005). Study of the Antarctic environment can also provide an insight into natural preindustrial aerosol processes, and allows us to further our understanding of the preindustrial baseline (Hamilton et al., 2014).

The role of aerosols in the Antarctic is poorly understood as their sources are many and varied in a relatively understudied region. Emissions from the Southern Ocean and the Antarctic region are characterized by multiple environmental systems including open ocean water, sea ice regions, and land, snow covered and snow-free. These regions are being affected by our changing climate (Chen et al., 2009). It has been long known that the seasonal cycle of Antarctic particle number concentrations has a strong seasonal cycle (Shaw 1988), leading to the inference that most of the Antarctic aerosol concentrations is linked with biological processes occurring in the surrounding oceans. It has been proposed that a large "pulse" over summer months arises from the upper Antarctic plateau (Ito et al., 1993; James, 1989), although particle number concentrations are much higher in coastal Antarctica. In other words, it is still debatable if the aerosols originate in the upper troposphere, being

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transported by the Antarctic drainage flow (James, 1989) to the coast by katabatic winds (Ito, 1993; Koponen et al., 2002; Fiebig et al., 2014; Hara et al., 2011; Järvinen et al., 2013; Humphries et al., 2015) or the marine pelagic (ice-free) and sympagic (ice-containing) boundary layer are a source of ultrafine particles (Herenz et al., 2019; Weller et al., 2011, 2015, 2018; Dall´Osto et al. 2017; Heintzenberg et al., 2000, 2023).

The Antarctic aerosol summer maximum concentrations (mainly ultrafine particles, <100 nm) may be largely explained by new particle formation (NPF) events, as reviewed by Kerminen et al. (2018). Super-micrometre aerosols (>1 µm) will mostly arise from primary sea spray (O'Dowd et al., 1997a, 1997b; Rankin and Wolff, 2003). The accumulation mode (100 nm – 1 µm) is a complex intermediary region with both primary and secondary sources related to their multitude of sources and long atmospheric lifetimes (Fossum et al., 2018; Gras and Keywood, 2017). The relative importance of these sources undergoes seasonal cycles related to meteorology and biological productivity.

The roles of secondary aerosols produced from biogenic sulfur mainly derived from the atmospheric oxidation of dimethylsulfide (DMS, a trace gas produced by marine plankton) and primary sea-spray aerosols in regulating cloudiness above the Southern Ocean is still a matter of debate (Meskhidze et al., 2006; Korhonen 2008, Asmi et al., 2010; Quin and Bates, 2011, Fossum 2018, Lachlan-Cope et al., 2020). The seasonal cycle of primary and secondary aerosol emissions from the SO affect also light scattering by aerosols over all Antarctica, including the upper plateau (Virkkula et al., 2022). A recent intensification in Antarctic aerosol measurement field campaigns is revealing that aerosol chemistry in the higher latitudes of Antarctica can be much more complex than two broad natural sources governing the aerosol populations: sea spray (primary, mostly composed of sea salt) and non-sea salt sulfate ($nssSO_4^{2-}$; secondary). For example, Paglione et al. (2023) stressed that various - not yet fully elucidated - sources and aerosol processes are controlling the Antarctic aerosol primary and secondary population, with the emissions from sympagic and pelagic ecosystems affecting the variability of submicron aerosol composition both in maritime areas as in inner Antarctic regions.

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McCoy et al. (2015) suggested that primary marine organic ultrafine aerosols are important in the SO region. Saliba et al. (2021) found that the large total organic fraction of particles <0.1 µm diameter may have important implications for CCN number concentrations and indirect radiative forcing over the Southern Ocean. Recently, Humphries et al., (2021) identified three main aerosol sources in the Southern Ocean: northern (40-45 S), mid-latitude (45-65 S) and Southern sector (65-70 S), with different mixture of continental and anthropogenic, primary and secondary aerosols depending on the studied region.

Recently, several long-term measurements (in the order of a year or longer) of aerosol particle number size distributions (PNSD) at a high time resolution allow investigation of the aerosol sources around the Antarctic continent. The PNSD is typically measured from ~10 – 1,000 nm, and in Antarctica is usually comprised of particles from secondary (NPF) and primary (blowing snow - BS, Sea Spray Aerosols - SSA) sources.

At King Sejong Station on the Antarctic Peninsula, (Kim et al., 2017; Park et al., 2023; Kim et al., 2019), Halley on the mainland coast (Lachlan-Cope et al., 2020) and Concordia/Dome C in the centre of the continent (Järvinen et al., 2013), PNSD measurements indicated NPF as a summertime and wintertime phenomenon. At Concordia/Dome C weak and rare but real NPF has been observed even in June, the darkest time of the year (Järvinen et al., 2013). Particle counts and CCN numbers at Macquarie Island and throughout the Southern Ocean undergo a summertime maximum (Humpries et al., 2023). The CCN concentration at the onset of NPF showed an average instantaneous increase of 44% or 11% across the whole period compared with the background concentration (Kim et al., 2019; Park et al., 2023). NPF therefore seems to modulate particle and CCN counts in the Antarctic.

At Halley, NPF events were shown to arise both from the sea ice marginal zone and the Antarctic plateau, indicating a marine and free tropospheric source (Lachlan-Cope et al., 2020). NPF events at King Sejong Station were found more frequently associated with air masses originating from the Bellingshausen Sea than those from the Weddell Sea, with a fraction of events being associated with sea ice (Park et al. 2023). This suggests the phytoplankton composition of the Bellingshausen Sea may be a source of NPF precursors (Jang et al., 2019). In Marambio, Quéléver et al.,

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(2022) reported important observations of neutral and charged aerosol precursor molecules and chemical cluster composition (qualitatively and quantitatively) - as well as air ions and aerosol particle number concentrations and size distributions - linked with local fauna (mainly penguins), further away from the island; and possibly to the metabolic activity of plankton and accumulation of pre- cursors (e.g., proteins) in the melting sea ice.

Hara et al. (2021) hypothesized that NPF mainly occurred in the Antarctic free troposphere during spring and fall, and in both the free troposphere and boundary layer during summer, based on aerosol number size distributions and their effect on cloud properties observed at Syowa Station. Concordia/Dome C 's observations reported the background particle number size distribution (PNSD) had its largest mode below 30 nm, suggesting these particles were produced in the upper atmosphere before entering continental Antarctica (Järvinen et al., 2013). However, also ground-level particle formation takes place on the upper plateau. Chen et al. (2017) reported NPF events that were observed at Concordia/Dome C by using a combination of an Air Ion Spectrometer (AIS) and a Differential Mobility Particle Sizer (DMPS). In several events particle formation and growth started fom the cluster ion size range of 0.9 – 1.9 nm which means that in these cases the formation took place at the ground level. But the compounds, mainly sulphuric acid that are needed for the NPF and growth are not emitted from the upper plateau, their sources are far away. NPF in Antarctica is therefore dependent on biogenic emissions from the ice-covered and ice-free regions of the ocean, with NPF occurring both at ground-level and in the colder upper atmosphere where particle formation rates can proceed more rapidly.

Previous analyses of PNSD at Halley using k-means cluster analysis has shown that wintertime PNSD was characterised by extremely low particle concentrations, with a bimodal size distribution appearing with a blowing snow or sea spray origin (Lachlan-Cope et al., 2020). The Antarctic wintertime at this site is therefore mostly devoid of secondary aerosol sources. NMR analyses of ambient aerosol samples show that aerosols arising from the ice-free Southern Ocean are rich in lipids and sugars, aerosols arising from the ice-covered Weddel sea are rich in methanesulphonic acid and amines, and aerosols arising from coastal areas are rich in sugars associated

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with plant vegetation (Decesari et al., 2020). The primary aerosol sources are therefore many and varied across Antarctica.

Here, we apply k-means cluster analysis to simultaneous measurements in the year 2015 at four Antarctic research sites, extending the study of Lachlan-Cope et al., (2020). We also compare the yearly data with field studies, including the PEGASO cruise in 2015 (Dall´Osto et al., 2017a, 2019b; Rinaldi et al., 2019; Decesari et al., 2020) and the continental Antarctic site Kohnen (Weller et al., 2018). We show a prevalence of new particle formation at the coastal sites, and associate this new particle formation with air masses flowing over regions of sea ice and ocean. At the more southerly and inland sites, secondary particles dominate the particle number concentrations, while air masses primarily travel over regions of land. Ambiguity remains in this analysis, as some PNSD clusters likely contains a substantial contribution from primary and secondary processes. Nonetheless, we provide further evidence for the roles of emissions from sympagic and pelagic ocean regions in new particle formation, and highlight the many and varied sources of particles across Antarctica.

2 Methodology

2.1 Sampling sites and measurements

Simultaneous PNSD measurements at four sites across Antarctica were collected for analysis. Their locations are stated in Table 1, and shown in Figure 1. Data coverage shown in Figure S1. PNSD measurements were aggregated to 1 hour for this study.

The South Korean King Sejong Station, the highest latitude site, is located in the Antarctic Peninsula. The PNSD from 10 to 300 nm were measured every 3 min with a mobility particle size spectrometer (MPSS) consisting of a differential mobility analyzer (DMA; HCT Inc., LDMA 4210) and a CPC (TSI 3772). Details of the site can be found in (Kim et al., 2017, 2019, Park et al., 2023).

The Argentinian Marambio Station is located in the Antarctic Peninsula, approximately 3 km from the coast. The PNSD from 12 – 800 nm were measured every 6 min with a differential MPSS (Asmi et al., 2010; Quelever et al., 2022).

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The French & Italian operated Concordia/Dome C station is operated on the East Antarctic Plateau at an elevation >3000 m a.s.l, and 900 km from the nearest coast. The Concordia/Dome C sampling site is located 1 km southwest of Concordia/Dome C's main building. The PNSD from 10 – 620 nm were measured every 10 min with a differential MPSS system (Järvinen et al., 2013).

The British Halley VI station is located in coastal mainland Antarctica on the floating Brunt Ice Shelf ~ 20 km from the coast of the Weddell Sea. The The Clean Air Sector Laboratory where PNSD measurements are taken is located 1 km south-east of the station. The PNSD from 6 to 209 nm is measured every 1 min using a TSI Inc. MPSS, comprising an Electrostatic Classifier (model 3082), a Condensation Particle Counter (CPC; model 3775) and a long Differential Mobility Analyser (DMA; model 3081) (Lachlan-Cope et al., 2020).

2.2 Data processing

k-means Cluster Analysis was applied to the PNSD data to apportion the PNSDs according to their shape (Beddows et al., 2009), and is routinely applied in pristine environments to PNSD data (Dall'Osto et al., 2017b, 2019b; Lachlan-Cope et al., 2020). k-means apportions data into k clusters such as the sum of squares of distances of data to the cluster centre is minimised. In the case of MPSS data, this produces well-separated clusters (Beddows et al., 2009). First, data were normalised so the Euclidian length of each PNSD was 1, so the clustering is done solely on the merit of the shape of the PNSDs. Here, a degree of distance between clusters was achieved with 16 MPSS clusters. These were assigned into 6 categories typical of Antarctic PNSDs (Lachlan-Cope et al., 2020).

The condensation sink (CS, s^{-1}) represents the rate at which a vapour phase molecule will collide with pre-existing particle surface, and was calculated from the PNSD data as follows (Kulmala et al., 2012):

$$CS = 2\pi D \sum_{d_p} \beta_{m,d_p} d_p N_{d_p} \quad (1)$$

Here, we presume the vapour phase molecule in question is sulphuric acid.

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2.3 Air mass trajectories

Air mass back trajectories were calculated using the HYSPLIT4 (Hybrid Single Particle Lagrangian Integrated Trajectory) trajectory model (Draxler and Rolph, 2010) arriving at the sampling site every one hour. Each back-trajectory data point was assigned to a surface-type (land, sea, ice, or snow over land. A cell is considered ice-covered if more than 40% of the cell is covered with ice) on a 24 km grid from the daily Interactive Multisensor Snow and Ice Mapping System (IMS) (Ice, 2008). To investigate air masses concurrent with high particle count, these 72-hour back-trajectories were gridded to 1x1 grid cells of 1 degree each, and linked back to the total integrated particle number concentration from the PNSD measurements by the following equation:

$$ln(\bar{C}_{ij}) = \frac{1}{\sum_{k=1}^{N} \tau_{ijk}} \sum_{k=1}^{N} \ln(c_k)\, \tau_{ijk} \quad (2)$$

Where $\bar{C}_{ij}$ is the concentrated weighted trajectory (CWT) at cell i, j, N is the total number of trajectories, c_k is the value of particle number (N) associated with the arrival of trajectory k, and τ_{ijk} is the residence time of trajectory k in grid cell i, j. $\bar{C}_{ij}$ therefore describes the source strength of condensable vapour that drives particle growth from any particular grid cell (Hsu et al., 2003; Lupu and Maenhaut, 2002). This was done using the trajLevel function in the Openair package in R 3.6.3.

3 Results

3.1 Trends in aerosol PNSDs

The mean PNSD for the measured periods are shown in Figure 2a. PNSDs at the two Antarctic Peninsula sites, Marambio and King Sejong show large nucleation modes. Aitken mode peaks are present at all sites with maximum concentrations between 30 and 50 nm. Accumulation mode peaks are present at all sites with maximum concentrations >100 nm. The prominent nucleation modes at Marambio and King Sejong are also visible in the daily contour plots, with the visible signature of a new nucleation mode in the late and early afternoons (local times) at the two sites, respectively (Figure S3). Total average particle counts are highest at King

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Sejong & Marambio, (312±601 and 270±541 cm^{-3}), particle counts at Halley are lower (223±245 cm^{-3}), while those at Concordia/Dome C Dome C are 44±67 cm^{-3}.

The monthly variation in total particle number (N_{tot}) is similar between all sites, highest in the austral summer, with minimum values in July & August. N_{tot} at Concordia/Dome C is substantially lower in near all months compared to all other sites, with the monthly average only exceeding 100 cm^{-3} in the Austral Summer months (Nov, Dec). Relative contributions of the modes <30 nm, 30-100 nm, and >100 nm to N_{tot} at each site is shown in Figure 2d. The relative contribution of <30 nm particles is highest at Marambio and King Sejong, exceeding 50% of the total N_{tot} at Marambio, consistent with the prominence of the presence of the nucleation mode (Fig. 2a). The contribution of <30 nm particles to total N_{tot} is lowest at Halley, where ~50% of the N_{tot} comes from the mode 30-100 nm, and ~30% from the mode >100 nm. At Concordia/Dome C, ~65% of N_{tot} comes from the mode 30-100 nm, with small contributions from particles above or below this size range.

3.2 Cluster analysis

3.2.1 Categorising PNSD K-means clusters

K-means cluster analysis was used to partition the PNSD data into days with similarly shaped PNSD data. In k-means cluster analysis, starting with a higher number of clusters and then merging them based on similarity can enhance the separation and interpretability of the final clusters by capturing finer details. These 16 daily K-means clusters (Figure S2, referred to onwards as the initial PNSD clusters) were therefore aggregated into 6 daily categories typical of Antarctic PNSD data (referred to as simply the PNSD clusters) for further analysis based on the shape of the PNSD and their temporal variation (Figure 3a and 3b). These are Nucleation, Bursting, Aitken low, Aitken high, Bimodal, and Pristine, following previous classification described in previous studies (Dall´Osto et al., 2017b; Dall´Osto et al., 2019, Lachlan-Cope et al., 2020). The diurnal cycle of the PNSD belonging to each of the initial 16 K-means clusters at each of these sites is shown in Figure S4, while

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the diurnal cycle of the merged clusters is shown in Figure S5, separated by site, and the PNSD per cluster separate by season is shown in Figure S6.

Two clusters relate to NPF: Nucleation (mean particle count 502±1087 cm^{-3}) has a mean PNSD with a peak at 12 nm averaged between all sites. This corresponds to fresh new particles generated during NPF events and will correspond to days of measurements dominated by a signature of new particles appearing in the PNSD, typically with signs of growth. Bursting (mean particle count 177±350 cm^{-3}) has a mean PNSD with a peak at 22 nm averaged between all sites, and typically corresponds to days where new particles form, either fail to grow, are lost to coagulation, or are diluted and lost at the receptor site. These likely still do correspond to secondary formation processes. We presume here that there is no primary contribution to these PNSDs

Aitken is the most common PNSD category separated into Aitken high (mean particle count 401.6±45.5 cm^{-3}), and Aitken low (mean particle count 208.8±216.7 cm^{-3}). The mean PNSD shows peak concentrations at about 20-35 nm and 35-55 nm respectively. These two clusters are separated based on their aerosol total concentrations. The name of this category emerges from growing ultrafine aerosol resulting from the processing of local and regional marine aerosols, a phenomenon previously described to occur mainly in summer (Dall´Osto et al., 2017b).

Pristine (mean particle count 142±134 cm^{-3}) has a mean PNSD with a peak at 88 nm. The name of this category emerges from the extremely low "pristine" particle number concentrations. The three K-means clusters belonging to this categories have some specific aerosol modes, peaking at 65 nm, 85 nm and a much larger accumulation mode at 160 nm (Figure S2) likely associated at pristine conditions with high loadings of blowing snow, as previously discussed in Lachlan-Cope et al. (2020).

Bimodal (mean particle count 107±133 cm^{-3}) has two peaks in the PNSD at 39 and 113 nm. Both are characterised by low particle counts. The name of this category is associated with the bimodal PNSD. The two initial clusters differ slightly because of the Hoppel minimum at 55nm and 75nm, respectively, possibly indicating different chemistry and particle origin.

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The condensation sink represents the main loss process for many low-volatility vapours which contribute to new particle formation. The condensation sinks for the Bursting K-means cluster is lowest ($2.3 \cdot 10^{-4}$ s^{-1}), followed by Bimodal ($2.4 \cdot 10^{-4}$ s^{-1}), Pristine ($3.4 \cdot 10^{-4}$ s^{-1}), Nucleation ($3.7 \cdot 10^{-4}$ s^{-1}), and Aitken low ($3.4\ 10^{-4}$ s^{-1}), and Aitken high ($4.0\ 10^{-4}$ s^{-1}). Particle counts change by around 1 order of magnitude between sites, but the shapes of the PNSD are similar (Figure S2, S5). All Nucleation PNSDs show formation of and growth of particles at midday and afternoon times (local time), while Bursting show particles at or near the smallest sizes, sometimes with indication of particle growth.

3.2.2 Trends in PNSD K-means clusters

The frequency of occurrence of these different PNSD clusters can indicate the dominant sources of aerosol particles at each of these time periods. All sites have a large contribution from Aitken low and Aitken high PNSD clusters, with 37% of PNSDs falling into these categories across all four sites. Marambio is also dominated by PNSD clusters falling into the Nucleation and Bursting category, (25% and 23% of PNSDs, respectively). There is a smaller contribution of Nucleation and Bursting to the PNSDs at King Sejong (8% and 20% respectively). These PNSD clusters contribute little at Halley (3% and 3%, respectively), and at Concordia/Dome C (2% and 5%, respectively). Halley instead sees a large contribution of Pristine PNSDs (45% of PNSDs), and Concordia/Dome C sees a large contribution of Bimodal PNSDs (34% of PNSDs) (Figure 3a).

The seasonal evolution of these K-means clusters is shown in Figure 3c. At Marambio, the contribution of Nucleation and Bursting PNSDs occurs in nearly all months. At King Sejong, Nucleation and Bursting PNSDs are most frequent in the spring and autumn months, while Pristine air masses are most common in the Austral winter. At Halley, Aitken PNSDs dominate the warmer months, while Pristine air masses dominate the colder winter months. At Concordia/Dome C, where data are available, Bimodal PNSDs dominate the winter, while Aitken dominates the warmer months. There is a substantial contribution of Nucleation and Bursting in the month of September also. When all sites are averaged, a clear seasonal cycle is

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seen, with pristine and bimodal PNSDs dominating in the coldest months, and a substantial contribution of Aitken in the warmer months. Nucleation is slightly higher in the warmer months (particularly January through March), while bursting PNSDs are higher outside of these months.

3.3 Air mass analysis

72 hour HYSPLIT back trajectories arriving at the four different measurement sites for every hour with PNSD data were calculated. The amount of time each of these air masses spent flowing over land, open ocean, marginal ice, or sea ice per K-means cluster across all sites is plotted in Figure 4a, and per site is plotted in Figure 4b. On average, the air masses arriving at the sites had flown over mostly the land of the Antarctic continent (54%), with a 24% contribution from sea ice, 15% contribution from open water, and a small (6%) contribution from marginal ice. Nucleation & Bursting PNSDs occur when air masses have travelled under substantially less land and over more sea ice, marginal ice, and ocean than average (40%, 8%, and 26% of air mass hours for Nucleation, and 34%, 10%, and 25% of air mass hours for Bursting, respectively). At Marambio, the main contribution is air masses arising from sea-ice regions (50%) which is highest for Bimodal PNSDs (76%). The contribution of open water to Aitken, Bursting & Nucleation is higher than average (23%), while the contribution of sea ice to these air masses is lower (44%). At King Sejong, air masses spend the majority of their time travelling over regions of open water (41%) and sea ice (33%). The contribution of open water is higher for bimodal (51%) and Aitken PNSDs (58%). At Halley, air masses spend the majority of their time flowing over land regions (79%), with a small contribution from sea ice (15%). The contribution of sea ice to Nucleation PNSDs is substantially higher (30%). At Concordia/Dome C, air masses arriving at the receptor site mostly flow over land (99%). The contribution of sea ice, marginal ice, and open water are higher for bursting PNSDs (9% in total).

Our CWT (concentrated weighted trajectory) analysis grids back trajectories to 1x1 degree squares and weighs each segment of the back trajectory with the corresponding N_{tot} observed upon the air mass's arrival, performed individually for each PNSD cluster. These are plotted in Figure 5. A map highlighting source regions for N_{tot} unseparated by cluster per site is shown in Figure S8. The CWTs separated

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by cluster and site is shown in Figure S9. Mean heights of these trajectories are shown in Figure S10.

The sources of particles by number (or, N_{tot}) across all PNSD data at the two Antarctic Peninsula sites Marambio and King Sejong are to the west of the peninsula, across the iced and open water regions of the Bellingshausen Sea and to a lesser extent the Weddell sea. Those at Halley are concentrated at the coastal and land-based regions, with some influence from the Weddell sea, while the highest aerosol concentrations at Concordia/Dome C arise from mostly land regions, with some trajectories extending past the southern tip of the continent over ocean regions (Figure S8).

Air masses arriving at the Antarctic sites corresponding to Nucleation & Bursting K-means clusters pass mostly over the Bellingshausen sea at Marambio and King Sejong. Air masses passing over the regions just west of the peninsula have a high source contribution to high number concentrations of particles especially, as do the more northerly regions off the southern tip of south America. At Halley, these PNSD clusters are mostly associated with coastal regions, consistent with the higher contribution of sea ice. The source contributions at Concordia/Dome C correspond to a small number of trajectories. All trajectories corresponding to Nucleation PNSDs have a lower-than-average trajectory height (Figure S10).

Aitken PNSDs have strong source contributions from the entire Bellingshausen sea region at Marambio and King Sejong. At Halley, there is a strong contribution from land mass regions, while at Concordia/Dome C, land and regions mostly contribute to this PNSD. Bimodal and Pristine air masses at Marambio and King Sejong have strong source contributions from the iced regions either side of the Antarctic peninsula, covering the Weddel and Bellingshausen seas. This is consistent with the higher contribution from sea-ice regions to these K-means clusters. At Halley, the Bimodal PNSDs arise when air masses flow over coastal and land-based regions, while Pristine PNSDs, which are extremely frequent at this site, arise largely from the Antarctic continent itself. At Concordia/Dome C, there is a strong source contribution to Bimodal & Pristine PNSDs from air masses flowing over the coastal region of southern Antarctica, as well as some land regions. Trajectories corresponding to Pristine PNSDs have, on average, a higher trajectory height than average.

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3.4 Intercomparison with other existing data across the year 2015

3.4.1 PEGASO cruise

The PEGASO (Plankton-derived Emissions of trace Gases and Aerosols in the Southern Ocean) cruise was conducted on board the RV Hesperides in the regions of Antarctic Peninsula, South Orkney and South Georgia Islands from 2 January to 11 February 2015. It was found that the microbiota of sea ice and the sea ice-influenced ocean can be a source of atmospheric primary and secondary organic nitrogen (ON), specifically low molecular weight alkylamines (Dall´Osto et al., 2017; 2019). Other follow up studies also claim that the potential impact of the sea ice (sympagic) planktonic ecosystem on aerosol composition were overlooked in past studies, and multiple eco-regions act as distinct aerosol sources around Antarctica (Decesari et al., 2020; Rinaldi et al., 2020).

Figure 6a shows the intercomparison of MPSS PNSDs for the PEGASO cruise. These are separated into times when the air masses flowed over the Pacific ocean, and over the Weddel sea. This is compared with stations used for this study where overlapping data are available (Marambio, King Sejong, and Halley). Air masses from the Southern Pacific Ocean with anthropogenic and terrestrial influence from Patagonia are likely responsible for the higher Aitken mode seen for the "Pacific" PNSD during the PEGASO cruise. By contrast, cruise measurements influenced by the Weddel sea show a PNSD similar to the three stations intercompared, suggesting little anthropogenic influence of the latter. Dall'Osto et al., (2017) showed that sea ice regions are a strong source of sub-3 nm particles in Antarctica relative to open water regions. The present study, considering a much broader dataset, also shows that the Open Water (OW) regions sampled at Marambio and at King Sejong stations are also associated with enhanced NPF.

3.4.2 Kohnen station

Weller et al., (2018) reported PNSDs and conducted bulk and size-segregated aerosol sampling during two summer campaigns in January 2015 and January 2016 at the continental Antarctic station Kohnen (Dronning Maud Land). The transport of aerosols was associated with two main weather conditions: (1) the sporadic impact

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of cyclones causing high loadings of marine aerosol concentrations and NPF and (2) clear sky conditions causing long range transport of aged aerosols. Figure 6b shows similar aerosol PNSDs among Kohnen, Halley and King Sejong stations for the same sampling period in 2015. By contrast, much higher concentrations of ultrafine particles - likely due to NPF - were seen occurring at Marambio. This may be a combination of local sources (nearby penguin colonies) and melting sea ice during the summer period, given Marambio is the station among all closest to marginal sea ice zones (Quelever et al., 2022).

4 Discussion

4.1 Possible primary sources

Of our six aerosol categories, nucleation and bursting are related to secondary aerosols, while the two Aitken K-means clusters likely have significant contributions from primary and secondary processes (Figure 2a), although the latter may dominate. Bimodal PNSDs are likely associated with aged and processed marine aerosols (the Hoppel minimum mode is seen at about 70 nm, Hoppel et al., 1994). We cannot apportion primary aerosols from our PNSD measurements without any chemical composition information, but can hypothesise some. Sea salt aerosol particles are formed from open ocean regions when sea spray is produced by wave breaking and bubble bursting generating film and jet drops (De Leeuw et al., 2011). Sea salt aerosol is also formed from blowing snow above sea ice (Frey et al., 2020). These mechanisms are thought to contribute equally to sea salt aerosol loads (Legrand et al., 2016), while blowing snow over land is a source from continental Antarctica (Giordano et al., 2018).

In the periods where NPF is negligible, there are still other primary aerosol sources. We show that pristine and bimodal PNSD clusters dominate through the winter (Figure 3c, S6). It is therefore important to characterize these pristine conditions. The three K-means clusters of the pristine categories have similar low particle number concentrations, but one of the three has a distinct PNSD. The cluster (pristine_160) seen with a bimodal PNSD (75 and 160 nm, respectively), was strongly associated with high wind speed and possibly associated with blowing snow and sea spray sea salt, dominating the winter aerosol population (Lachlan-Cope et al., 2020). When the

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four stations are compared in this study, it can clearly be seen that Halley has the most frequent pristine conditions (45%, relative to the 12-20% of the other three), also relative to Concordia/Dome C where we propose that free troposphere transport and aged aerosols populate the PNSD with Aitken modes and bimodal PNSDs.

The pristine initial PNSD cluster with a strong peak at 160nm (table S1) is seen mainly at Halley (12%) relative to the other stations (2-3%). A study by Yang et al. (2019) proposes a source for ultrafine sea salt aerosol particles from blowing snow, dependent on snow salinity. This mechanism could account for the small particles seen during Antarctic winter at coastal stations (Giordano et al., 2018; Frey et al., 2020). Recently, similar PNSD were reported by Gong et al., (2023) in the central Arctic over an entire year from September 2019 to October 2020 during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) expedition, showing that blowing snow was observed more than 20% of the time from November to April. The sublimation of blowing snow generates high concentrations of fine-mode sea salt aerosol (diameter below 300 nm), enhancing cloud condensation nuclei concentrations up to tenfold above background levels (Gong et al., 2023).

Primary marine organic aerosol in the submicron regime is likely produced separately from sea salt (Gantt and Meskhidze, 2013). At the coastal sites of Marambio and Halley, Bimodal and Pristine PNSDs with the highest particle number concentration arise mostly from the Bellingshausen Sea, where DMS(P)-rich phytoplankton concentrations are very high. A dominant biological oceanic mechanism of primary particle origin at these sites is therefore likely. Given the vastly different source characteristics between these sites, it is likely that the dominant mechanisms of origin of these primary particles, and therefore our bimodal and pristine clusters also differ between sites, but it is clear that primary emissions dominate the PNSD in the Antarctic winter.

4.2 New particle formation and possible secondary sources

We identify two PNSD clusters which we classify as arising from secondary processes, these are Nucleation and Bursting PNSDs. The Aitken PNSD clusters cannot be definitively said to contain solely particles from secondary processes,

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although they have similar source regions (Figure S5) and could consist of grown particles formed either elsewhere in the boundary layer or in the free troposphere, with average growth rates around ~0.1 – 1 nm h^{-1} (Brean et al., 2021; Järvinen et al., 2013; Jokinen et al., 2018; Kim et al., 2019), particles formed through NPF would grow to the sizes observed in this PNSD cluster on the order of ~2 days. The contribution of Aitken PNSDs is greatest outside of the winter months (Figure S7). However, below, we will purely discuss the contribution of the Bursting and Nucleation PNSDs.

NPF in Antarctica is a summertime phenomenon largely responsible for the seasonal cycle in particle number concentrations (Figure 2c). This cycle has been observed at at King Sejong Station (Kim et al., 2017; Park et al., 2023; Kim et al., 2019), Halley (Lachlan-Cope et al., 2020) and Concordia/Dome C (Järvinen et al., 2013; Chen et al., 2017). This corresponds with periods of high biological and photochemical activity (Jang et al., 2019; Kim et al., 2019). These observations are consistent with our observed higher frequency of Nucleation PNSD clusters in January through March (Figure 3c, S6). However, we consistently observe Nucleation and Bursting PNSDs year-round, even at sites where manual inspection of these PNSDs has found little to no NPF in the winter seasons (Kim et al., 2019).

We frequently observe PNDSs with a large nucleation mode in the Austral summer (Figure S7). NPF is mainly influenced by the source rates of vapours (DMS, VOC, base, and iodine emissions, $OH^{\cdot}$ and O_3 production rates), loss rates of vapours and new particles (CS), ion pair production rates, and temperature (Lee et al., 2019). Vapour source rates will be lowest in winter, however, ion pair production from cosmic rays is likely constant, while loss rates of vapours and temperatures have a wintertime maximum. Some number of particles may therefore form and be identified by Cluster analysis, even when they do not give the visual signature of an NPF event.

Our analyses show that coastal Antarctic sites nearest to the melting sea ice are most influenced by NPF (Figure 3c) where a large fraction of the PNSDs are classified as Nucleation and Bursting (48 and 28% at Marambio and King Sejong respectively). Further, we show that NPF is frequent when air masses flow over the ice-influenced oceanic Antarctic regions (Figure 4,5). NPF events have been shown

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to be frequent and strong at coastal Antarctic sites (Brean et al., 2021; Jokinen et al., 2018), but most rapid at those nearest to melting ice (Brean et al., 2021) as well as near local sources associated with penguin colonies (Quelever et al., 2022). Recent observations shows that this does not extend across the whole of the southern ocean (Baccarini et al., 2021). Our PNSD cluster analysis is consistent with this. Melting sea is therefore a likely source of NPF precursors in the Antarctic Peninsula region.

High aerosol number concentrations arising from the Bellingshausen sea is consistent with a previous analysis of NPF at King Sejong (Jang et al., 2019). Bursting and nucleation aerosol categories in particular exhibit high number concentrations when air masses pass over the regions just west of the peninsula (Figure 5). These regions have been shown to be rich in alkylamines (Dall'Osto et al., 2019a) that lead to rapid and efficient particle formation (Brean et al., 2021). There is evidence that these melting sea-ice regions are sources of numerous NPF precursors (Atkinson et al., 2012; Dall'Osto et al., 2017a, 2019a). This is also consistent in the sympagic Arctic environment (Dall'Osto et al., 2017b, 2018). Consistent with this, the results of our K-means cluster analysis show that the sites with the highest exposure to air masses that pass over regions of sea ice and marginal ice have by far the greatest contribution from nucleation and bursting PNSD types (Figure 2, 4). The sites where air masses primarily travel over snow-covered land regions have, by contrast, very little contribution of nucleation and bursting (Figure 4a).

The role of new particles formed in the free-troposphere and transported down to our receptor sites is unknown, but likely. Particle formation processes are highly efficient in the free troposphere given the lower temperatures and higher ion pair production rates. A wider number of potential nucleation mechanisms are therefore likely (Kirkby et al., 2011; Wang et al., 2022). Modelling studies predict a substantial fraction of CCN in all seasons (Korhonen et al., 2008). These particles, if arriving at the receptor site long enough after formation, will contribute either to Nucleation, Bursting, or Aitken PNSDs. While we therefore highlight source regions leading to boundary layer NPF processes, we cannot state what fraction of PNSDs arise from free tropospheric particle formation processes.

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During the austral summer the concentration of DMS in the water of the Southern Ocean is the highest of the planet (Lana et al., 2011), with high fluxes into the atmosphere, potentially producing high concentrations of sulphuric acid and methanesulphonic acid. However, under typical boundary layer conditions the concentration of sulphuric acid is too low to form particles alone, and another molecule, such as ammonia, is required to stabilize the nucleating clusters (Kirkby et al., 2011). Jokinen et al. (2018) reported the first molecular characterization of NPF from the Aboa station in Antarctica, showing that new particles are formed via clustering of sulfuric acid and ammonia (Kirby et al., 2011). Sources of ammonia and alkylamines are related to animals, mainly birds and seal colonies and melting sea ice, but the relative importance of each source is unknown (Riddick et al., 2012; Schmale et al., 2013, Brean et al., 2021; Quelever et al., 2022). Iodine oxoacids are also capable of efficiently forming new particles (Baccarini et al., 2020; He et al., 2021; Sipilä et al., 2016), with iodous acid being able to efficiently stabilise clusters of iodic and sulphuric acid, even in the absence of ammonia (He et al., 2023), with modelling studies showing high gas-phase iodine concentrations across Antarctica (Saiz-Lopez et al., 2007). Further, the formation of marine primary aerosol particles and of gas-to-particles precursors is influenced by the uppermost layer of the ocean, the so called sea surface microlayer (SML) (Cunliffe et al., 2013). The SML - with physicochemical characteristics different from those of subsurface waters (SSW) - results in dense and active viral and microbial communities (Vaque et al., 2021, Heinrichs et al., 2024). The microlayer of the ocean is a source of oxygenated VOCs in the Arctic leading to secondary aerosol formation (Mungall et al., 2017), and it is also likely a source in the Southern Ocean too although no measurements of this type are available.

Therefore, while the limited number of measurement studies implicate sulphuric acid and nitrogenous bases as the drivers of NPF, iodine and VOCs may play a substantial role.

5 Implication and conclusion

To gain insight into the influence of marine Antarctic biogeochemistry on atmospheric aerosol, we report simultaneous aerosol PNSDs collected across an entire year (2015) at four research stations (Marambio, King Sejong, Halley, and

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Concordia/Dome C), as well as a brief intercomparison with a cruise around the regions of Antarctic Peninsula, South Orkney, and South Georgia Islands (Dall´Osto et al., 2017) and a field campaign in Kohnen station (Weller et al., 2018). Our study shows that the aerosol PNSDs across the Antarctic have striking differences, confirming that multiple eco-regions, with subsequent atmospheric chemical and physical processes, act as multiple aerosol sources around Antarctica. These analyses suggest that the PNSD of Antarctic sub-micrometre aerosols may have been oversimplified in the past (Ito, 1993) and that complex interactions between multiple ecosystems, coupled with different atmospheric circulation, result in very different PNSDs populating Antarctica. Our knowledge on aerosol sources of primary and secondary origin is limited.

Recently, Brean et al., (2023) emphasized how understanding the geographical variation in surface types across the Arctic is key to understanding secondary aerosol sources, highlighting that particle formation and growth rate vary tremendously likely due to different regions producing different precursors source rates. We argue that the same complexity applies to Antarctica - an ensemble of regions with substantial spatial heterogeneity across marine, terrestrial, and freshwater biomes, with productivity and biodiversity patchiness super-imposed on strong environmental gradients. Antarctica, one of the world's eight major biogeographical realms, is made up almost entirely of one single bioregion - the ice-covered land mass, coastal tundra, and sea ice of the main continent. It contains two additional marine bioregions - the Antarctic Peninsula & Scotia Sea and the Subantarctic Indian Ocean Islands (Chown et al., 2007; Terauds et al., 2012, 2016). A bioregion is smaller in scale than a biogeographical realm but larger than an eco-region or an ecosystem, it allows the integrations of multiple eco-regions, including terrestrial, freshwater and marine into a cohesive system. Whilst these definitions may be challenging in the context of atmospheric biogeochemistry, we argue a better understanding of the interactions between the biosphere and the geosphere are needed to better understand aerosol sources in Antarctica.

Antarctic sea ice covers between 1% (summer) and 5% (winter) of the global ocean. Antarctic terrestrial productivity and biodiversity occur almost exclusively in ice-free areas that cover less than 1% of the continent. In Antarctica the coastline extends for

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17,968 km and comprises about 34.8-36.4x10^6 km^2 where 80% of this surface is covered by ice, even in summer (Peck et al., 2018; Ronowicz et al., 2019). Overall, there is about 390,071 km^2 of coast shallower than 200 m (Peck et al., 2018). Antarctic coastal systems harbour a high diversity of marine and terrestrial ecosystems including Antarctic seaweeds (benthonic macroalgae) and bird colonies (mainly penguins). Antarctic seaweeds (often called macroalgae) are found in shallow (<200m) coastal, rocky shores and can cover more than 80% of the benthic surface (Amsler et al., 2005; Wiencke and Amsler, 2012). The Antarctic and Sub-Antarctic region is home for about half of the total worldwide seabird population (Otero et al., 2018). Penguins represents a high proportion of the avian biomass, and their fecal material is one of the main source of phosphorus and nitrogen, representing about 80% of these elements in the Antarctic marine environment. Seabird colonies also represent a significant source of atmospheric ammonia (NH_3) in remote maritime systems (Riddick et al., 2012, Schmale et al., 2013). These emissions are environmentally relevant as they primarily occur as “hot-spots” in otherwise pristine environments.

Figure 7 (a, b) shows a schematic illustration of the sea ice, microbiota, sea-to-air emissions, and primary and secondary aerosols in Antarctica. The Southern Ocean is among the largest source of Sea Spray Aerosols (SSA) on our planet Earth. Current aerosol models have a large uncertainty in the SSA abundance (Lapere et al., 2023) and the relative importance of the sublimation of blowing snow is not yet quantified (Giordano et al., 2018; Frey et al., 2020). The biogenic organic component of SSA in Antarctica is thought to be important (Mc Coy et al., 2015) but again not fully quantified. Sea ice may also modulates SSA production, with potentially significant climate impacts (Dall´Osto et al., 2022a). Other leached material from Antarctic media including seaweeds and penguin guano may also affect cloud-relevant aerosol SSA production (Dall´Osto et al., 2022b).

The changes occurring in the Antarctic environment will modify the climate with feedbacks and exchanges between the biosphere and cryosphere with the atmosphere. Changes in marine and terrestrial life - including adaptation of ecophysiology, food and nutrient availability - will affect the emissions of primary and secondary aerosol precursors. Interdisciplinary studies and international cooperation

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in Antarctica are reducing the gap in our knowledge of these key environmental factors.

Code and data availability.

The code and data used to produce all non-illustrative figures are available from the corresponding authors under reasonable request.

Supplement.

The supplement related to this article is available online at: X

Author contributions.

Conceptualization: MDO conceptualized the paper following the study of Lachlan-Cope et al (2020).
Data Curation: EA provided data for Marambio; TLC and AJ provided data for Halley, AL, AV, PA provided data for Concordia/Dome C, and JP and YJY provided data for King Sejong.
Software: DCSB and JB.
Formal Analysis: DCSB, JB, MDO.
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Project Administration: MDO.
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Visualization: JB.
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Competing interests.

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Site	Lat	Lon	Elevation a.s.l. (m)
King Sejong	-62.2	-58.8	10
Marambio	-64.2	-56.6	198
Concordia/Dome C	-75.1	123.3	3233
Halley	-75.6	-26.2	30

Table 1: Latitude & longitude of each site

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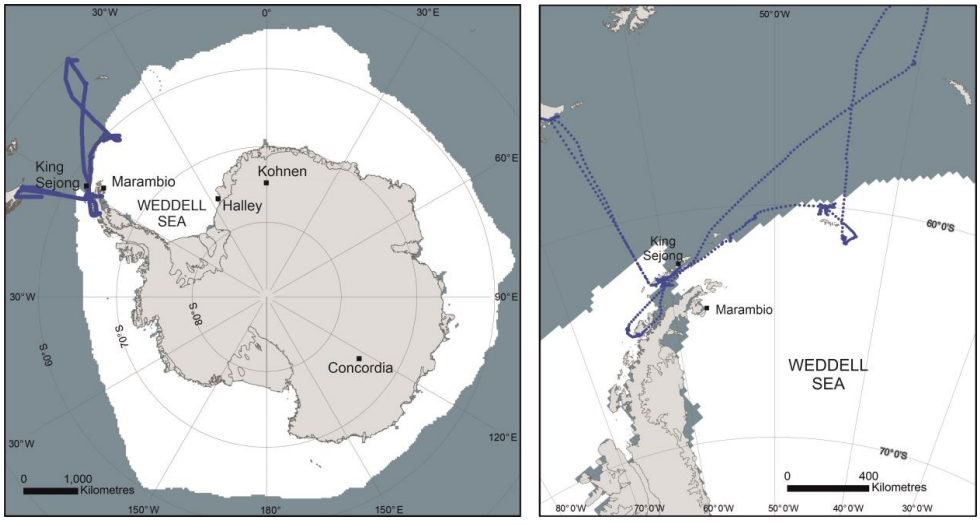

Figure 1 Map of the sampling stations (Halley, Marambio, Concordia/Dome C, King Sejong) for the year dataset collected in 2015. Additional, data for shorter period are intercompared at Kohnen (Weller et al., 2018) and during the PEGASO cruise (Dall´Osto et al., 2017, blue line the PEGASO cruise track). Please note that the sea ice extent is the median September extent from 1981 to 2010 (data are from the National Snow and Ice Data Center – NSIDC – at https://nsidc.org/data/, last access: 17 March 2024, Fetterer et al., 2017).

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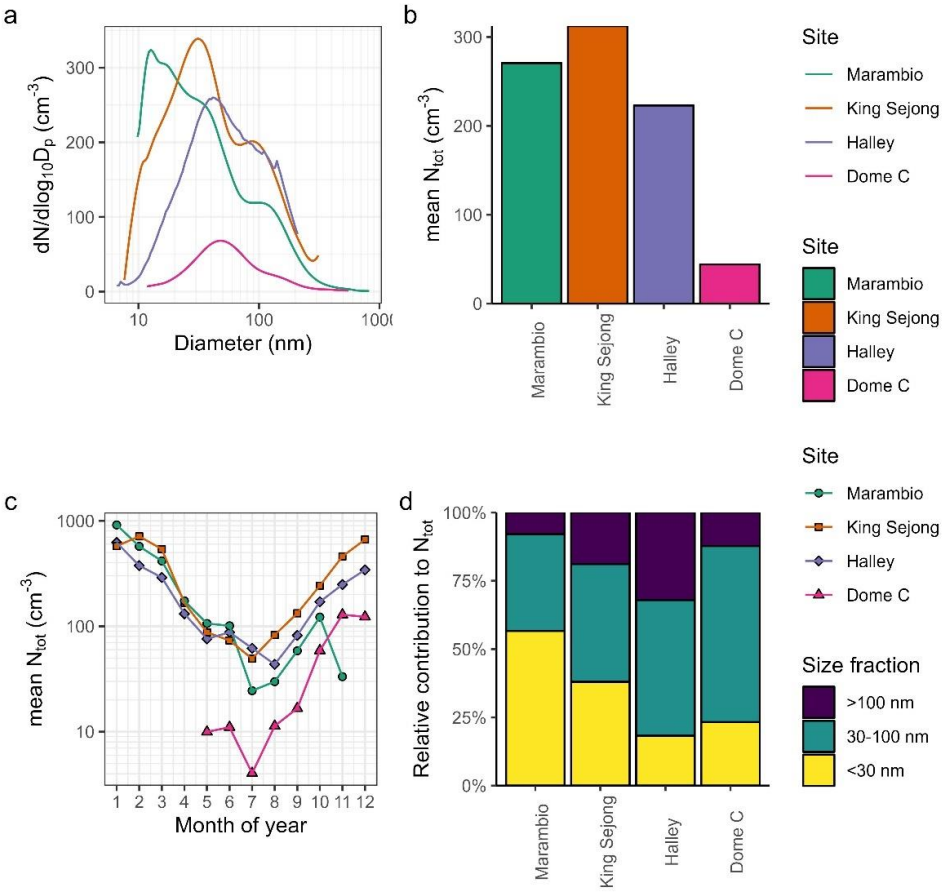

Figure 2: Features of particle PNSD data for four Antarctic sites, showing (a) mean particle PNSD per site, (b) mean particle count per site, (c) seasonal variation in particle count per site, and (d) contribution of different size fractions to particle count per site. Only data from the same time period (April-December) are intercompared and presented in Figure 2a,b,d, whereas all the temporal trend is presented in Figure 2c.

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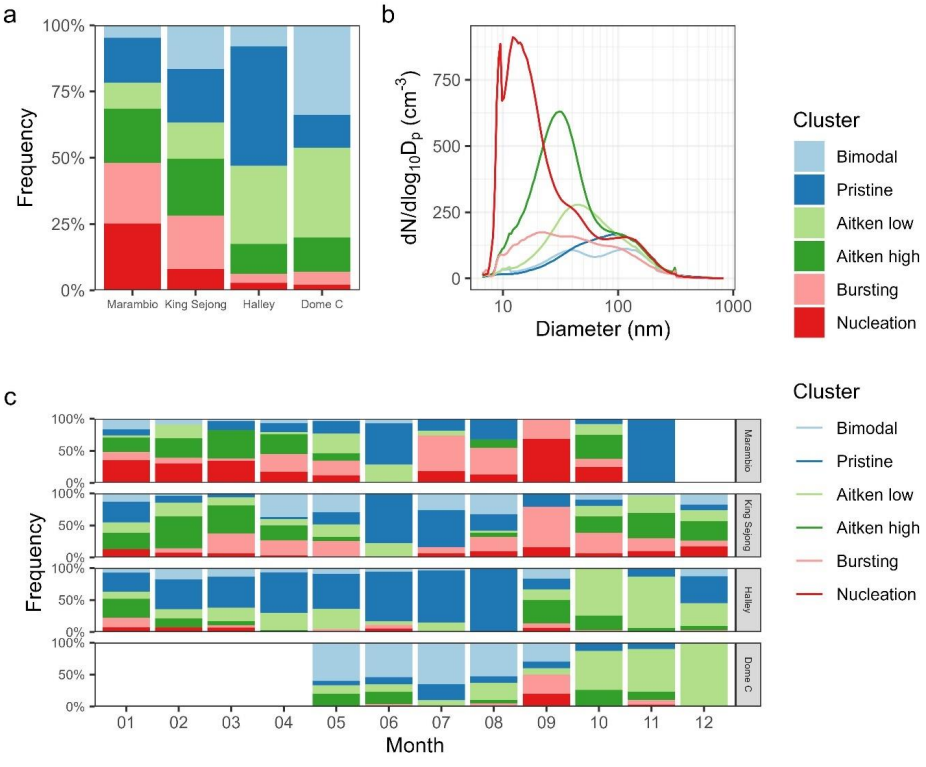

Figure 3: Cluster analysis results, showing (a) average frequency of each cluster per site, (b) mean PNSD per cluster, and (c) seasonal variations in cluster frequency per site

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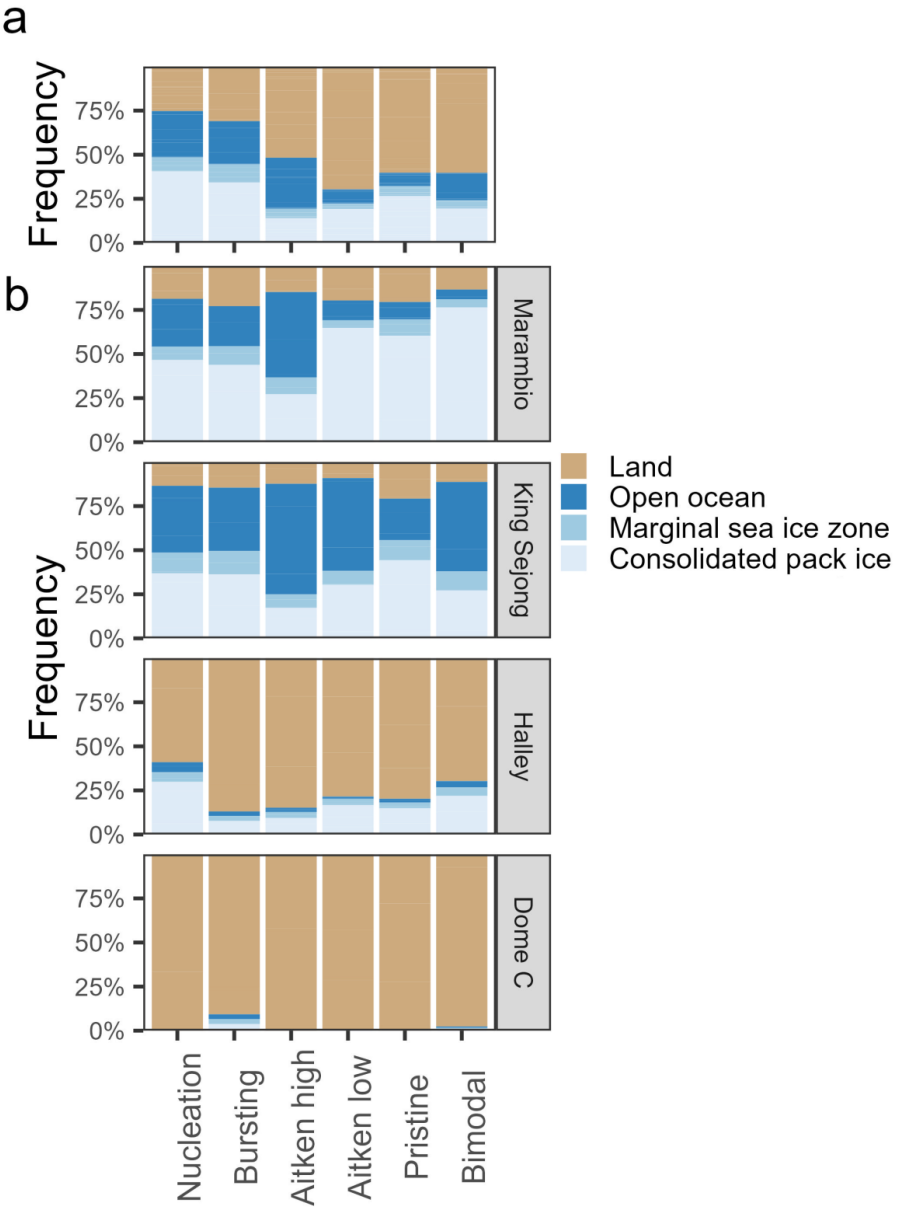

Figure 4: Land surface types associated with each cluster, showing (a) average association across all sites, and (b) association per site

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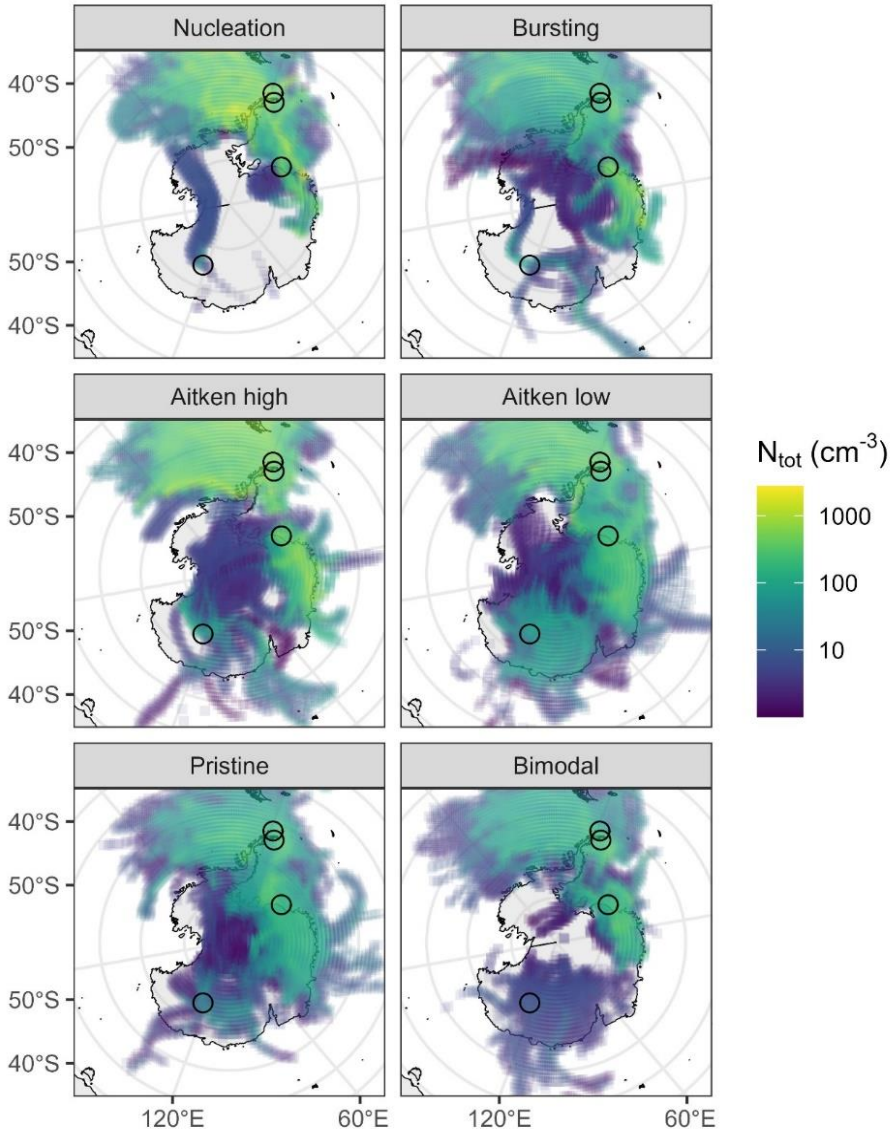

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Figure 5: Concentration weighted trajectory maps showing the sources of particles corresponding to each cluster. 72 hour back trajectories calculated with Hysplit

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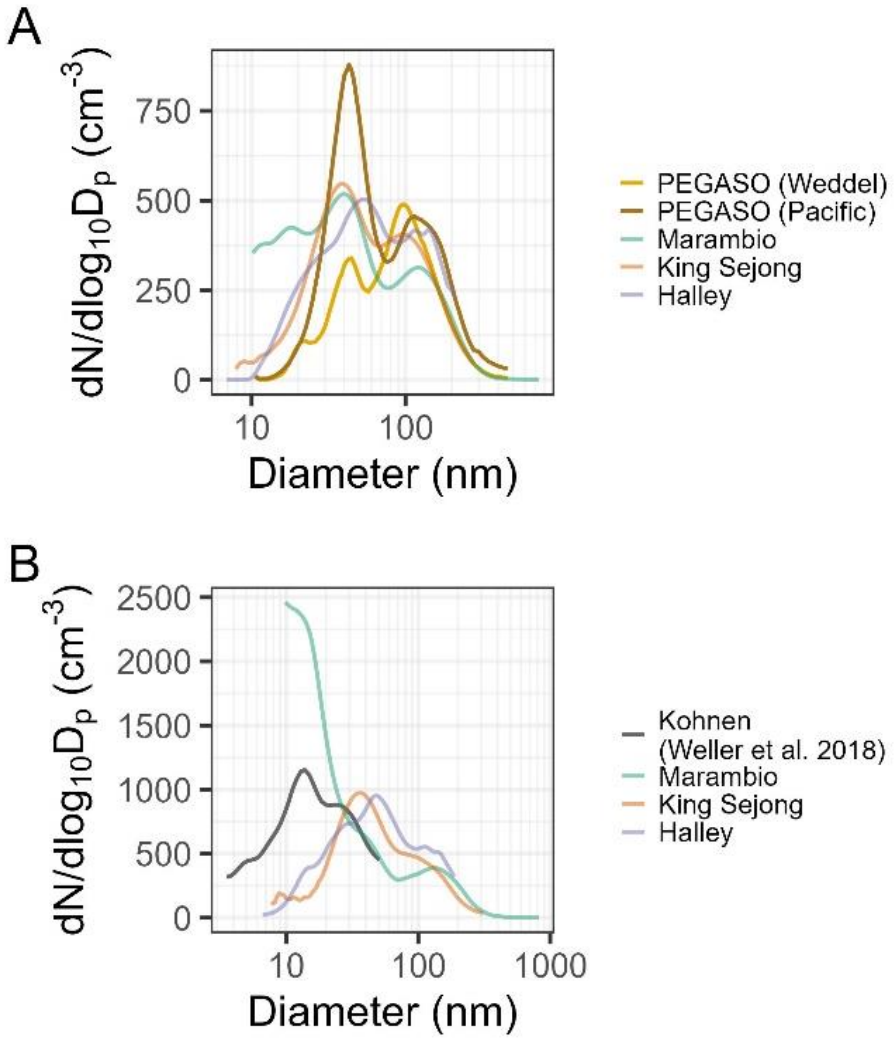

Figure 6: PNSD intercomparisons: (A) PNSD from the PEGASO cruise both when influenced from air masses from the Weddel Sea and Pacific Ocean and the stations used for this study where overlapping data are available (Marambio, Halley and King Sejong stations), and (B) PNSD from the Kohnen station (Weller et al., 2018) and the stations used for this study where overlapping data are available (Marambio, Halley and King Sejong stations)

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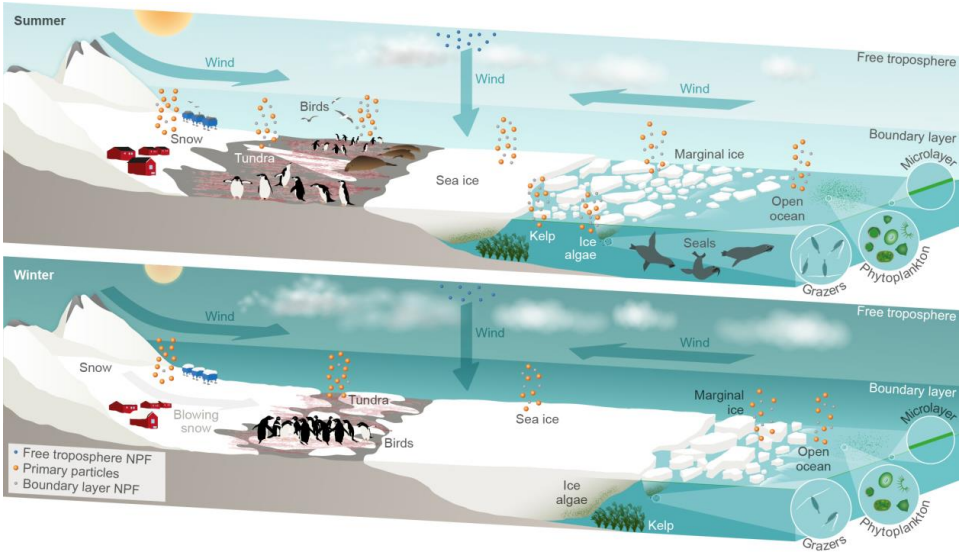

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Figure 7 Schematic illustrations of the sea ice, microbiota, sea-to-air emissions, and primary and secondary aerosols in Antarctica during summer (top) and winter (bottom).