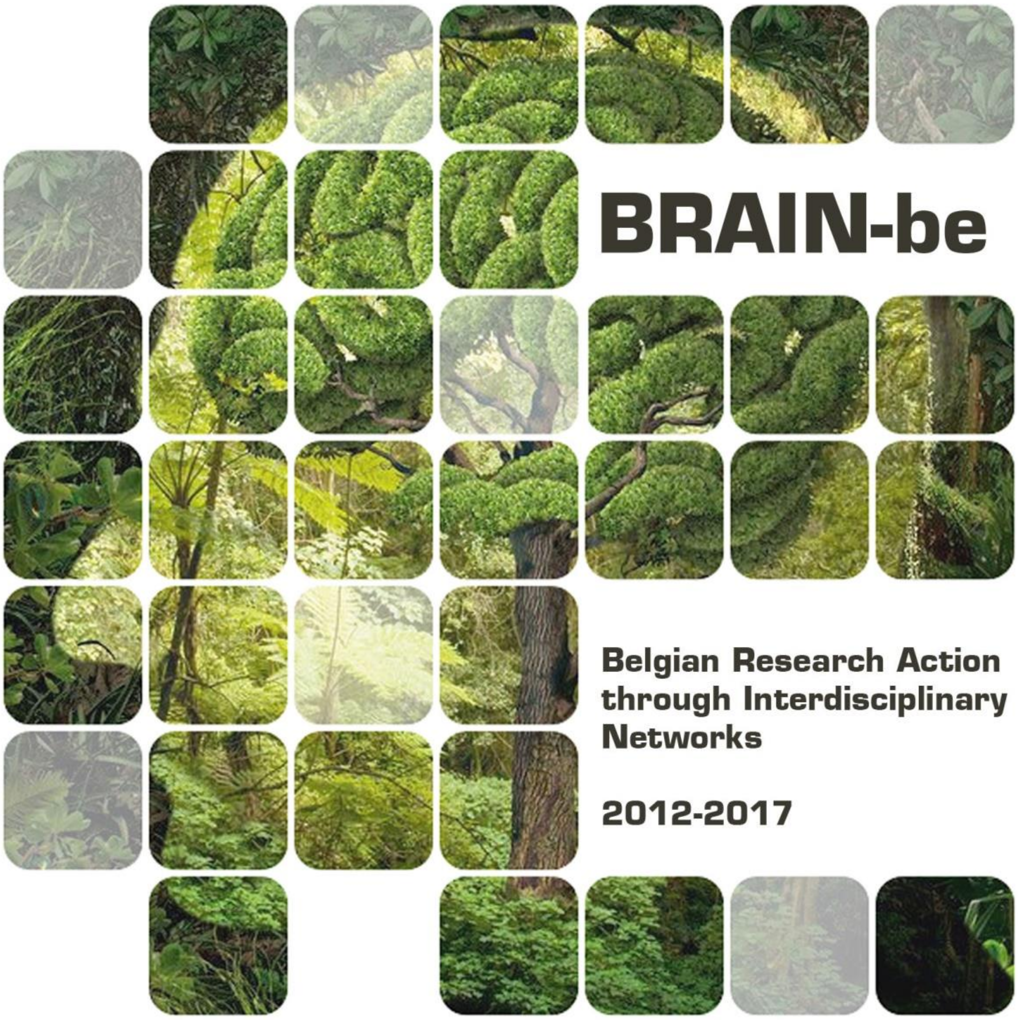

CHASE

Unravelling Particle Chemistry in Dronning Maud Land: from Atmosphere to Surface

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NETWORK PROJECT

CHASE

Unravelling Particle Chemistry in Dronning Maud Land: from Atmosphere to Surface

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FINAL REPORT

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ABSTRACT

Context

The fluxes and sources of atmospheric particles and gaseous compounds in Antarctica and its closely associated Southern Ocean are poorly constrained, in particular the particle chemistry. Antarctica is considered the best preserved region on Earth from anthropogenic emissions. However, the impact of anthropogenic airborne particles and pollutants could be significantly larger than expected. Furthermore, a detailed understanding of present-day atmospheric transport pathways of particles and of (semi-)volatile organic compounds ((S)-VOCs) from source to deposition in Antarctica remains essential to document biogeochemical cycles and the relative importance of natural and anthropogenic compounds, which are not well constrained at the moment. In addition, atmospheric particles also act as cloud condensation nuclei and ice nuclei and therefore play a crucial role in the formation of clouds, affecting both their radiative properties and precipitation.

The CHASE project provided detailed physical-chemical analyses of both atmospheric and particles in surface snow as well as of (semi)-volatile organic compounds recovered near the Belgian research station Princess Elisabeth (PEA), Dronning Maud Land, East Antarctica (71.95°S, 23.35°E, 1390 m asl), and thoroughly investigated their atmospheric transport pathways. Such detailed studies have never occurred in the region where Princess Elisabeth station is located.

CHASE relied on an interdisciplinary research team, bringing together partners with complementary expertise in long-term experimental work, expertise in Antarctic research campaigns, expertise in state-of-the-art chemical analyses, and expertise in modelling atmospheric transport and dispersion. Both active and passive sampling of ambient atmospheric particles and of (semi)-volatile organic compounds has been done. Active sampling allowed a higher time resolution of the samples during austral summer. Passive sampling allowed to obtain time-weighted average concentrations over longer periods of time and offering the possibility to sample in remote areas. The location of the passive sampling sites along a transect from the Antarctic plateau to the coast allowed the collection of samples influenced by various source regions.

Objectives

The objectives of CHASE were:

- To build up a unique database of organic and inorganic composition of both atmospheric and surface snow particles as well as of volatile organic compounds in Dronning Maud Land, East Antarctica.
- To assess comprehensively source regions, atmospheric transport pathways, seasonal variations in Dronning Maud Land, East Antarctica.
- To improve the understanding how the Antarctic atmospheric composition is influenced by lower latitudes.
- To valorise the existing observatory at Princess Elisabeth station and to set up towards the end of the project a long-term monitoring of the organic and inorganic atmospheric chemistry by passive sampling in the vicinity of PE station.

Conclusions

- The amount of polycyclic aromatic hydrocarbons (PAHs) and oxygenated PAHs in the particle phase were found to be negligible. Very likely this was due to the very low atmospheric particle number concentration measured at Princess Elisabeth station. Fluorene, phenanthrene, fluoranthene and pyrene were the most ubiquitous PAH compounds found in the samples with concentrations ranging between 1 and over 100 pg/m³. No significant inter-annual differences were found for these compounds.
- 158 samples for the analysis of volatile organic compounds (VOCs) were collected in which around 65 compounds were identified and if possible quantified. This resulted in a dataset of over 10000 data points making it the largest dataset on VOCs and oxygenated VOCs in Antarctica. It can be concluded that oxygenated aromatic compounds are by far the most important group by concentration. Acetophenone, phenol, benzaldehyde and benzoic acid are known oxidation products of primary aromatic compounds and are present in concentrations up to 2 µg/m³. Further, dimethylsulfone ($DMSO_2$), an oxidation product of dimethylsulfide (DMS), clearly showed a decreasing trend in function of the distance of the sample site to the ocean.
- For the first time, carbon isotope ratios of particulate organic carbon (POC) and dissolved organic carbon (DOC) were determined in surface snow samples in the region of PEA. The linear correlation between the DOC-flux and ssNa+-flux indicated that sea spray was the main source of DOC. This was confirmed by the carbon isotopic ratio of the DOC.
- The inorganic chemical analyses showed that the large majority, up to 89 %, of the sampled particles were below 2 µm and up to 50 % of particles are of submicron size. Practically no particles with a size > 5 µm were detected. This particle size pattern showed no significant distinction along the 250 km measurement transect from coast to the plateau, neither when comparing air to surface snow samples.
- For the first time, samples were collected for the analysis on the concentration of ice nucleating particles (INP) for the region of Dronning Maud Land. Compared to studies in other regions of Antarctica, the INP numbers for PEA are at the lower limit. This is an important finding, particularly for modelling studies on the aerosol influence on cloud formation and precipitation.
- The entire sample set presented a comparable mineralogical composition dominated by aluminosilicate, silica and Mg-Fe silicates closely followed by Fe-bearing aluminosilicates and iron or titanium oxides. In a much lesser proportion and non-systematically, metal-bearing particles composed of Cr, Ni, Zn, Cu, Sb, Sn, Tl, Ta were present, indicating anthropogenic sources. Special attention was given to particles containing iron (Fe), as Fe is a key micronutrient, essential for the primary productivity in the austral ocean. The occurrence of Fe-bearing particles was found to be widespread in East Antarctica.
- From chemical and isotopic analyses of particles deposited in surface snow, a novel statistical model based on the found patterns of rare earth elements (REE) has been developed. An additional major potential source area (PSA) for dust particles in East Antarctica could be

identified. Besides confirming that Southern South America is the best candidate to explain the dust signature recorded during cold and warm geological periods, this study proposes for warm periods a scheme with also Southern Africa as PSA.

- A climatology of backward air mass trajectories has been established for the first time for the region of East Antarctica around PEA, covering a period of 11 years (2010-2020). A k-means cluster analysis has been performed and four clusters of air mass origin were found. Source regions from South America, Southern Africa and Australia were found to be very limited. The Southern Ocean was a main source region, as was the Antarctic continent itself. For the most important air mass cluster, the source region is mostly restricted to the region above the Antarctic continent and the average altitude along the trajectories in this cluster indicated that this cluster corresponded mainly to air subsiding from the upper troposphere.

These findings clearly demonstrate the value of the inter-disciplinary approach of the CHASE project, combining high-end analytical techniques, innovative sampling methodologies, expertise in atmospheric transport modelling, and in Antarctic field expeditions, in order to unravel the complex atmospheric chemistry in remote areas under harsh meteorological conditions. Although much progress has been made, some gaps have been identified that need to be addressed in future research. First of all, in order to discriminate potential source regions even better, more samples would be required, covering several years and with higher temporal resolution during winter. Ideally such measurements would take place both near the coast as well as on the Antarctic plateau at the same time, as CHASE proved that samples of these areas showed distinctly different chemical features. Moreover, the chemical fingerprints found by the CHASE analyses, clearly link certain source regions (e.g., Southern Africa) or other anthropogenic sources (chemical patterns of PAH, VOC, metals) to the sample locations in East Antarctica. But respective simulations of current atmospheric transport models have large uncertainties when simulating many weeks of atmospheric transport what apparently is necessary. Therefore, more elucidated modelling and input from measurements would be required to disentangle how these compounds from lower latitudes reach East Antarctica. Finally, it is recommended to continue with at least the collection via passive sampling at one or two of the CHASE sampling sites in order to extent these valuable time series. In order to gain a higher temporal resolution also during austral winter and to be independent of the potential contamination by the research station operation, low-flow automatic sampling systems with zero-emission power generation under harsh conditions need to be developed.

Keywords

Atmospheric particles / Atmospheric transport modelling / East Antarctica / Elemental and isotopic composition / Volatile organic compounds (VOCs)

1. INTRODUCTION

The fluxes and sources of atmospheric particles and gaseous compounds in Antarctica and its closely associated Southern Ocean are poorly constrained, in particular the particle chemistry. Antarctica is considered the best preserved region on Earth from anthropogenic emissions. However, the impact of anthropogenic airborne particles and pollutants could be significantly larger than expected. Furthermore, a detailed understanding of present-day atmospheric transport pathways of particles and of (semi-)volatile organic compounds ((S)-VOCs) from source to deposition in Antarctica remains essential to document biogeochemical cycles and the relative importance of natural and anthropogenic compounds, which are not well constrained at the moment. This information is however relevant to interpret climatic data extracted from ice cores and the transport and deposition of not only mineral nutrients, but also and essentially of organic micro-pollutants in polar regions.

Further, atmospheric composition change is a main driver of present and near-future climate change with atmospheric particles playing a major role therein. The impact of atmospheric particles as a major source of micronutrients (e.g. iron) in the so-called “High Nutrient Low Chlorophyll” oceanic zones (like the Southern Ocean) that affects the global CO_2 cycling, illustrates the close and complex relationships between the composition of atmospheric particles and global climate. In addition, atmospheric particles also act as cloud condensation nuclei and ice nuclei and therefore play a crucial role in the formation of clouds, affecting both their radiative properties and precipitation.

The CHASE project provided detailed physical-chemical analyses of both atmospheric and surface snow particles as well as of (semi)-volatile organic compounds recovered near the Belgian research station Princess Elisabeth (PEA), Dronning Maud Land, East Antarctica (71.95°S, 23.35°E, 1390 m asl), and thoroughly investigated their atmospheric transport pathways. Such detailed studies have never occurred in the region where Princess Elisabeth station is located.

CHASE relied on an interdisciplinary research team, bringing together partners with complementary expertise in long-term experimental work, expertise in Antarctic research campaigns, expertise in state-of-the-art chemical analyses, and expertise in modelling atmospheric transport and dispersion. Both active and passive sampling of ambient atmospheric particles and of (semi)-volatile organic compounds has been done. Active sampling via high volume pumps was essential to gain sufficient mass for the laboratory analyses. Active sampling allowed a higher time resolution of the samples during austral summer. Passive sampling allowed to obtain time-weighted average concentrations over longer periods of time at relatively low cost, requiring no electricity or sampling pumps, and offering the possibility to sample in remote areas. The location of the passive sampling sites along a transect from the Antarctic plateau to the coast allowed the collection of samples influenced by various source regions.

2. STATE OF THE ART AND OBJECTIVES

2.1 Characterisation of the organic atmospheric composition (particulate matter and (semi-)volatile organic compounds)

2.1.1 Organic composition of particulate matter and semi-volatile organic compounds (SVOCs)

The largest fraction of the limited research on semi-volatile organic compounds (SVOCs) in Antarctica has been focussing on historical pollutants like PCBs and pesticides such as DDT. These compounds end up in the Polar Region via the global distillation and cold trapping mechanisms and are important to monitor as the high bioaccumulation potential poses a threat for local wildlife. Since the Stockholm protocol the emission of PCBs and pesticides worldwide plummeted, but they remain present in the environment. Polycyclic aromatic hydrocarbons (PAHs) are a class of carcinogenic and persistent organic pollutants (POP) and are made up of only carbon and hydrogen atoms forming concatenated aromatic ring structures. PAHs are emitted during various combustion processes and anthropogenic activities and are distributed all over the globe via atmospheric transport (Ravindra et al., 2008). The unique conditions of polar regions (low temperatures, remoteness, strong light seasonality) imposes different behaviour for PAHs compared to temperate regions (Drotikova et al., 2020) but this has never been measured in-situ. Atmospheric transformation of PAHs occurs via the reaction with the hydroxyl radical (•OH) and ozone (O_3) forming oxy-PAHs, which are regarded as more toxic than the parent compounds. Some studies (n=4) measured PAHs in Antarctica, the most comprehensive probably being (Piazza et al., 2013) at the Mario Zucchelli Station where PAHs were measured during the summer of 2009 and 2010 using a high volume sampler (HVS). Nitro-PAHs were measured by (Vincenti et al., 2001) but to our knowledge oxy-PAHs were never measured in Antarctica. We aim to construct a long time-series of PAHs and oxy-PAHs by on-site measurements using an HVS in order to elucidate atmospheric transport to this specific area of Antarctica and reactions. The measurement will only take place during summer because of the electrical power required and the need for weekly manual sample changes.

Moving to the more volatile fraction of atmospheric organic compounds, VOCs, and their atmospheric oxidation products (OVOCs) play an important role in the atmosphere and global climate processes. Most VOCs have an atmospheric lifetime of a few hours to a several weeks, with halogenated compounds being a notable exception. They are readily transformed to compounds with a lower volatility by atmospheric oxidation and were never expected to reach Antarctica in large quantities. The atmospheric chemistry in the polar regions is very distinct as there are some specific conditions such as the polar day and night, powerful polar vortices, and strong UV radiation which are not relevant anywhere else in the world (Slusher et al., 2010). Despite this, the number of research publications dealing with the presence and concentration levels of VOCs is scarce. Compounds of the chlorofluorocarbon (CFC) group are among the most frequently monitored gases in the Antarctic atmosphere because of their relevance in the destruction of stratospheric ozone. But CFCs are not very relevant in the chemistry of the lower atmosphere, in particular the troposphere. Other VOCs on the other hand interact with different climate processes, e.g., the hydrological cycle and radiative forcing and these mechanisms form a big knowledge gap in our current understanding of the climate. The current knowledge on these compounds in the polar regions and especially

Antarctica is very limited. Several studies (Beyersdorf et al., 2010; Koga et al., 2014; Preunkert et al., 2007; Read et al., 2008; Swanson et al., 2004) measured dimethylsulfide (DMS), which is emitted from the ocean and the most important contributor to organic sulphur in the atmosphere, and oxidation products. Ethane and other low molecular weight hydrocarbons are reported in a few studies (Beyersdorf et al., 2010; Clarkson and Martin, 1997; Kaspers et al., 2004; Rudolph et al., 1989; Rudolph et al., 1992). To our knowledge, only Ciccioli et al. (1996) published data dedicated to oxygenated VOCs (OVOCs) in Antarctica. A total of 76 different VOCs were identified at 6 sample sites located near Terra Nova Bay proving the ubiquitous occurrence of these compounds. It can be concluded that a big knowledge gap exists on atmospheric VOCs in East-Antarctica, which play an important role in atmospheric processes.

2.1.2 Concentration, distribution and sources of organic carbon in East Antarctic snow

The significant fraction of carbon contained in the polar ice sheets constitutes an integral component of the global carbon cycle. The organic carbon composition and cycling in Antarctica is barely understood. Warming and changes in ice sheets and glaciers could affect global carbon dynamics (Antony et al., 2014), however, studies on organic carbon remain sparse in the Antarctic. In the framework of CHASE, the VUB carried out a preliminary study of the particulate and dissolved organic carbon concentration in surface snow samples of East Antarctica, along a transect from the coast to the plateau in the vicinity of Princess Elisabeth Station. The carbon sources were estimated using ion concentrations and carbon isotopic composition.

2.2 Characterisation of the inorganic composition of atmospheric particles

Mineral dust is considered a sensitive and powerful climatic proxy that allows the reconstruction of atmospheric circulations in the present or the past, and thus helps modellers to predict the future climate. Dust is also a sensitive natural tracer of modern and past environmental conditions at the deposition site but also at the emission site during the key climatic periods and their transitions. Introduced by Delmonte et al. (2004a), the term Potential Source Areas (PSAs) denotes either a primary source of mineral dust, derived directly from the physical and/or the chemical weathering of the parent material or a secondary source for dust, where airborne particles have already been subjected to a phase of aeolian and/or fluvial transport. Therefore, the type and intensity of dust emissions are a consequence of different conditions at the source (sediment supply and availability) and aeolian processes (deflation, transport, deposition) (e.g., Shao et al., 2010); dust remains in suspension until physical and chemical conditions favour its deposition. In Antarctica, most of the studies on the mineral dust have been carried out on ice cores extracted from the East Antarctic plateau (e.g., EDC, EDM, Vostok, Taylor Dome, Berkner Island). These works have mainly focused on the determination of the dust origin, based on Sr, Nd and Pb isotopic compositions (Delmonte et al., 2008; Vallelonga et al., 2010), rare earth and trace elements (Gaiero et al., 2004; Gabrielli et al., 2005), particle induced X-ray emission (Marino et al., 2008), magnetic properties (Lanci et al., 2008), or single element analysis (Siggaard-Andersen et al., 2007).

For the Southern Hemisphere, Southern South America (SSA), Australia (AUS), New Zealand (NZ), Southern Africa (SAF), and the Antarctic Dry Valleys have all been identified as PSAs. The first study to trace the dust origin in Antarctica ice cores was published by Grousset et al. (1992). In agreement with the Sr-Nd isotope data presented in that study, all the studies since have demonstrated that the main dust contributor to the Southern Ocean and East Antarctica during glacial (MIS2-MIS20) and interglacial periods is SSA, mainly from Patagonia (e.g., Basile et al., 1997; Delmonte et al., 2004a; 2019; Gaiero, 2007; Vallelonga et al., 2010; Gili et al., 2016, 2017). Despite the agreement on the dominance of a Patagonian dust origin during glacial periods, there is currently a debate regarding the Holocene dust PSAs with the following scenarios of dust provenance proposed: *i*) a weakened Patagonian source complemented by a lower latitude source region (Southern Central West Argentina, SCWA) (Gaiero, 2007; Gaiero et al., 2004; Gili et al., 2017); *ii*) a Puna-Altiplano dust source transported via the southern tropical jet stream (Gaiero, 2007; Gaiero et al., 2013; Gili et al., 2016, 2017) and, *iii*) a general contribution from Southern South America plus a secondary input from AUS (Revel-Rolland et al., 2006; Marino et al., 2008; De Deckker et al., 2010; Gabrielli et al., 2010;Du et al., 2018) or NZ (Wegner et al., 2012). Some of these geochemical and mineralogical interpretations were partially supported by Atmospheric General Circulation Models (AGCM), suggesting that while SSA dust dominates deposition over the Atlantic Southern Ocean and East Antarctica, AUS modern dust prevails over the southern Pacific and towards the western coastal regions of Antarctica (e.g., Li et al., 2008; Krinner et al., 2010; Albani et al., 2012). Additionally, two important studies (De Deckker 2019, 2020) reviewed and evaluated the role of AUS as a dust source during modern and Late Quaternary times showing that Australian dust likely reached certain sectors of Antarctica, such as Taldice and Taylor Glacier, predominantly during interglacial periods. For NZ, back-trajectory modelling showed that the transportation of aeolian material from NZ to the southern high latitudes is possible (Neff and Bertler, 2015). However, a recent study carried out on sediments from the major present-day dust and sediment producing regions of the South Island in NZ, has indicated this area was not a significant dust supplier to East Antarctica (Koffman et al., 2021). Using AGCMs, Li et al. (2008) showed that after SSA and AUS, Southern Africa is the third main dust supplier in the Southern Hemisphere with dust depositions mainly taking place in the South Atlantic and Indian Oceans. Even though this region is a significant dust emission source, little is still known about it being a PSA as the geochemical and modelling data available for this region is extremely sparse.

Consequently, there are still many unknowns about the origin and long-distance transport of the dust that reached Antarctica. This can be explained by the extreme conditions encountered during the sampling campaigns and also by the extremely low concentrations of trace elements, involving real challenges in the field and in the laboratory. The ice core records indicated that dust fluxes were up to 25 times higher during glacial periods due to dust production strengthening at their source areas, compared to warmer periods when dust concentrations were extremely low (Lambert et al., 2008). Therefore, the current interglacial period induces very challenging analytical measurements.

2.3 Air mass origin and transport pathways of atmospheric particles and compounds to East Antarctica

Antarctica is located far from anthropogenic activities and is one of the most pristine areas on Earth (Hamilton et al., 2014). In addition, the Antarctic region is sensitive to climate change. A changing environment in the Antarctic region will lead to changing sources and pathways of atmospheric particles. Respective measurements are therefore important in order to detect and to understand these changes. Limited aerosol sources are present on the Antarctic continent like dust from mountain areas (Virkkula et al., 2009), bacteria (Gonzales-Toril et al., 2009) and melt water ponds leading to local new particle formation (Kyrö et al., 2013). The Antarctic baseline aerosol budget recently has been found to originate from air masses of the free troposphere or lower stratosphere region, descending over the central Antarctic continent (Fiebig et al., 2014).

Another potential transport pathway of atmospheric compounds to Antarctica is by large-scale airflows which are directed towards the continent (e.g., Hara et al., 2010). Mineral dust can be advected to Western Antarctica within three to four days after emission from Southern America (Johnson et al., 2011). Along coastal Antarctica, natural aerosol sources can be very active. Due to the strong and persistent westerlies over the Southern Ocean, the mechanical source of sea salt particles is dominating particulate mass in the regional marine boundary layer (Jiang et al., 2021). East Antarctica is far more distanced from other mid-latitude continents and it is therefore essential to investigate pathways to that region.

Precursor gases for new particle formation (NPF) can originate from the Southern Ocean (e.g., dimethylsulfide, DMS) and possibly also from other sources, e.g., microbiota from sea ice and the ocean influenced by sea ice (Dall'Osto et al., 2017). Aerosols play an important role for cloud formation because they can act as cloud condensation nuclei (CCN) or ice nuclei (INP). Some studies have reported on Antarctic CCN properties, however, the locations they cover are limited (DeFelice et al., 1997; O'Shea et al., 2017). Within the last years new aerosol data have been accumulated, including the first Antarctic Circumnavigation Expedition directed at the pristine aerosol and also on CCN (Schmale et al., 2019). Further, it has been found that the role of changes in INP for clouds dominates over the role of changes in CCN (Solomon et al., 2018). About INP in the Antarctic region even less is known than on CCN. Ardon-Dryer et al. (2011) determined INP concentrations at the South Pole, where concentrations of 1 per Liter were found in the temperature range between -20°C and -24°C. Recently, Tatzelt et al. (2022) derived an extensive data set on CCN and INP. The study of SVOCs within CHASE yields information on potential precursor gases which can then via condensational growth or photolytic reactions add to the atmospheric budget of CCN and INP.

2.4 Objectives

The objectives of CHASE were:

- To build up a unique database of organic and inorganic composition of both atmospheric and surface snow particles as well as of volatile organic compounds in Dronning Maud Land, East Antarctica.
- To assess comprehensively source regions, atmospheric transport pathways, seasonal variations in Dronning Maud Land, East Antarctica.
- To improve the understanding how the Antarctic atmospheric composition is influenced by lower latitudes.
- To valorise the existing observatory at Princess Elisabeth station and to set up towards the end of the project a long-term monitoring of the organic and inorganic atmospheric chemistry by passive sampling in the vicinity of PE station.

3. METHODOLOGY

CHASE relied on an interdisciplinary research team, bringing together partners with complementary expertise in long-term experimental work, expertise in Antarctic research campaigns, and expertise in state-of-the-art chemical analyses. This complementary expertise of the different research groups and their collaboration led to a thorough investigation of the physical and chemical properties of the particles, the airborne pollutants in gaseous phase and their atmospheric transport which would never have been possible by the individual research groups alone.

Princess Elisabeth station (coordinates: 71.95°S, 23.35°E, 1390 m asl) station is excellently located for the studies of the CHASE project. It is 180 km inland from the coast, in the Sør Rondane mountains. Both access to the coast and also to the inland Antarctic plateau is fast (half a day on snow mobiles) and it is influenced by both synoptic and katabatic meteorological regimes (Gorodetskaya et al. 2013). Therefore, with measurements in the area of PEA, various source regions for atmospheric particles can be traced back. Note, however, that PEA is an austral summer-only station, open from mid-November to end of February.

Both active and passive sampling of ambient atmospheric particles has been done. Active sampling via high volume pumps was essential to gain sufficient mass for the inorganic and organic laboratory analyses. Active sampling allowed a higher time resolution of the samples during austral summer (not during austral winter, as PEA is not inhabited during that time period). Passive sampling allowed to obtain time-weighted average concentrations over longer periods of time at relatively low cost, requiring no electricity or sampling pumps, and offering the possibility to sample in remote areas. The location of the passive sampling sites along a transect from the Antarctic plateau to the coast allowed the collection of samples influenced by various source regions. Table 1 gives the coordinates of the sample locations and Figure 1 shows them on a map of Dronning Maud Land.

Table 1: Samples sites investigated by CHASE. At the end of the CHASE project, station Deep Plateau has been moved to the CLIMB site, used within the BRAIN-be CLIMB project; station Princess Elisabeth has been moved by 500m northeast due to snow accumulation and necessary lifting of the measurement pole

Sampling site	Latitudes	Longitudes	Altitude (m asl)
Breid Bay	70.30485 °S	23.61642 °E	75m
Frank Kenny North	70.43281 °S	23.84089 °E	110 m
Frank Kenny South	70.82900 °S	23.73500 °E	320 m
Romnoes	71.34678 °S	23.61131 °E	700 m
Princess Elisabeth	71.96014 °S	23.47353 °E	1320 m
Plateau	72.25336 °S	23.23195 °E	2300 m
Deep Plateau	72.37655 °S	23.41896 °E	2370 m
CLIMB site	72.27101 °S	23.25238 °E	2350 m

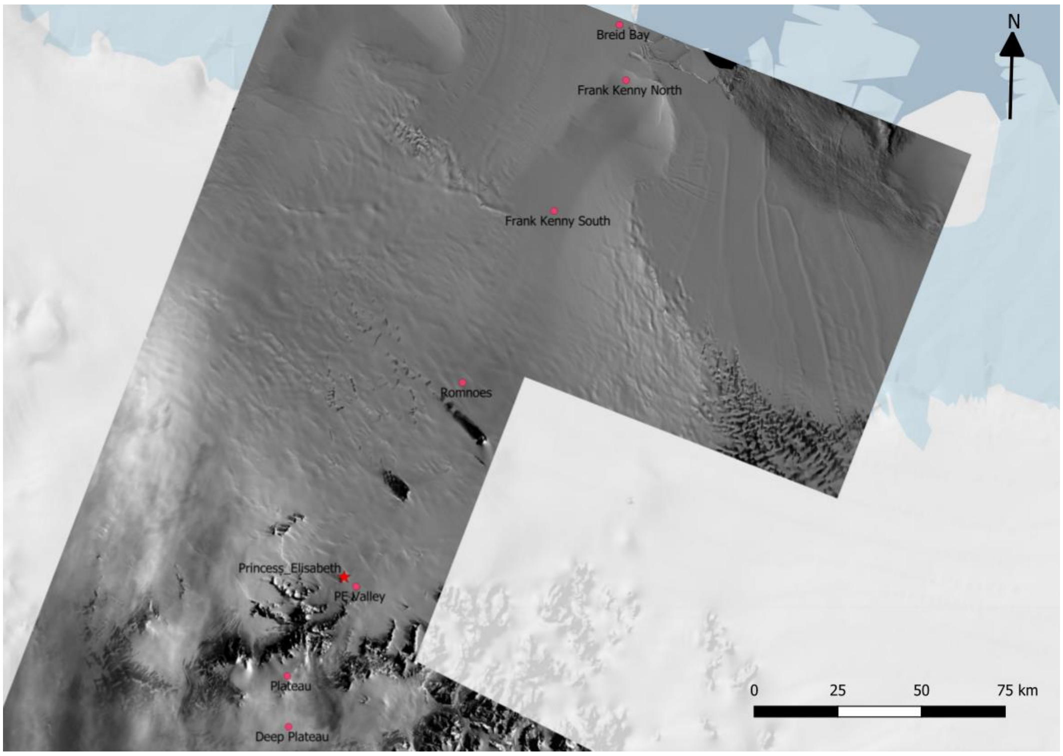

Figure 1: spatial location of the seven sampling sites where the CHASE passive samplers have been installed and where surface snow samples have been collected. 'Deep plateau', southward of PE, on the plateau; ii) 'Plateau', southward of PE, vicinity of the plateau; iii) 'PE valley', around 4 km eastward of PE; iv) 'Romnoes'; v) 'Frank Kenny South', between Romnoes mountains and the coast, vi) 'Frank Kenny North', near the coast; and vii) 'Breid Bay', near the coastline

3.1 Characterisation of the organic atmospheric composition (particulate matter and (semi-)volatile organic compounds

3.1.1 Active sampling of semi-volatile organic compounds (SVOCs)

To sample SVOCs both in the particulate and gas phase a Digitel DHA-80 high volume sampler (HVS, 500L/min) equipped with a holder for both quartz fibre filters (QFF) and a polyurethane foam filter (PUF was used. The instrument was installed in a stainless-steel plated measurement container 350 metres north of Princess Elisabeth Station. A PM_{10} impactor head was used, which cuts off all particles with an aerodynamic diameter above 10 µm. From previous research (BELSPO BRAIN project BR/143/A2/AEROCLOUD; www.aerocloud.be) it is established that atmospheric particles above this size are virtually not occurring in this area and the head mainly avoids intrusion of snow particles in the sampler. Due to the power requirement of a HVS, the instrument was powered directly from the station's supply. An ultrasonic anemometer was used to control the instrument in such a way that only air is sampled when wind is blowing from the "clean air sector", thus minimizing the sampling of local emissions. Pressure and temperature sensors were also installed, and the data logged to correct for the lower atmospheric pressure and temperature when expressing the SVOC concentration as mass per normalized volume unit.

Special care was taken during the method development to investigate the best approach to pre-clean both types of filters before sampling and how to transport them there and back while maintaining sufficiently low background levels. Each season 2 blanks were used as a quality control. Extraction of the samples took place with pressurized liquid extraction (PLE) using an ASE350 by Dionex with a mixture of acetone and n-hexane. This technique has the advantage of a reduced extraction time while maintaining the same extraction efficiency when compared to traditional Soxhlet extraction but highly reducing solvent usage, fitting in the green chemistry philosophy. The extraction efficiency of the used technique is expressed as analyte recovery and determined using sample material spiked with a standard mixture of the target compounds. To verify the recovery of PAHs adsorbed on particulate matter (PM) and caught by the QFF, urban dust reference material (NIST) was used. After solvent volume reduction the solvent in which the sample is dissolved is exchanged to toluene as it is compatible with the analytical instrument. Samples were finally analysed for 16 polycyclic aromatic hydrocarbons (PAHs) and 12 oxygenated PAHs (oxy-PAHs) on a GC-HRMS (Thermo, Finnegan MAT95XP-Trap, R=10000).

3.1.2 Passive sampling of semi-volatile organic compounds (SVOCs)

Passive sampling is noticeable different from the active sampling described in section **Error! Reference source not found.** in the way analyte is collected on the sampling substrate, driven by diffusion. No power is required for these devices to operate and the lower cost, when compared to HVS, made it feasible to install multiple of these devices on the transect from the Antarctic plateau south of the Sor Rondane mountains to the Princess Ragnhild coast to gain a spatial resolution in the SVOC data. From 2017 to 2021 passive air samplers (PAS) were installed at between 5 and 7 locations each time in duplicate. The sample devices were deployed for 1 year at a time to obtain a sample volume which is comparable with the HVS samples (average $2000m^3$).

Two different devices were investigated for the passive sampling of VOCs. Polydimethylsiloxane (PDMS) is frequently used in sample preparation for SVOCs in environmental samples and is applied as sampling medium as well. Thin PDMS sheets have been successfully deployed for sampling PAHs in marine environments and in other form factors for sampling SVOCs in air. Two 70 mm x 70 mm x 1 mm PDMS sheets were stacked on a custom-made quick release holder. A more established technique for sampling SVOCs in air is the usage of PUF disks which are exposed to the environment in a specific shelter made for passive sampling. The PDMS sampler was deployed during the 2017-2018 and 2018-2019 sampling campaign. The PUF type sampler (Tisch environmental TE-200-PAS) was deployed during all 4 sampling campaigns and transferred to the sites used by the BELSPO BRAIN-be CLIMB (B2/191/P1/CLIMB; https://ozone.meteo.be/projects/climb) project for 2 extra campaigns. Both types of samples were extracted using the ASE350 after developing and validating the extraction and clean-up method using reference material. Analysis was performed on a GC-HRMS (Thermo, Finnegan MAT95XP-Trap, R=10000).

3.1.3 Passive sampling of volatile organic compounds (VOCs)

VOCs were passively sampled on axial sorbent tube type sampler (stainless steel, 200 mg Tenax TA, Markes, UK). A shelter for the tubes was made from stainless steel together with an aluminum holder for 8 tubes. The shelter was installed 2m above the snow surface. Sorbent tubes were exposed for 1 year after which they were collected. The samples were analysed using a thermal desorber (Unity 2, Markes) coupled to a GC-MS (Thermo Scientific) and a novel technique which couples a PTR-TOFMS (high resolution) to a thermal desorber. A method was developed and validated to remove water from the samples.

3.1.4 Concentration, distribution and sources of organic carbon in East Antarctic snow and stable isotopes C and N of organic fraction

Partner VUB carried out a preliminary study of the particulate and dissolved organic carbon concentrations in surface snow samples of East Antarctica, along a transect from the coast to the plateau. The carbon sources were estimated using ion concentrations and carbon isotopic composition. Analysed samples come from the sites investigated by CHASE (see Table 1), during the field expeditions in austral seasons 2017-18 and 2019-20.

For the field campaign in 2017, snow samples from Frank Kenny North and South and Princess Elizabeth station were analysed and for 2019 Breid Bay, Frank Kenny South, Romnoes, Princess Elizabeth station, Plateau and Deep Plateau, forming a transect from coast to inland for 2019. There was no specific sample collection for this project. The samples analysed for C concentrations and isotopic signature are the remaining fraction of the sampled collected during the two campaigns, after other measurements were carried out.

Particulate organic C (POC) and N

For the filtration, Whatman GF/F pore size 0,7µm and diameter 47mm were used. The snow was melted over night at room temperature. Glassware was cleaned by leaving it in an acid bath for several hours. Glassware, filters and silver cups were baked in the oven at 450°C for 4h to remove all possible contamination. The same glassware was used for the different samples. Between each sample the glassware was rinsed five times with MilliQ water. The filtered volume varied between 785 and 975mL. The filter was cut into smaller pieces. Material used for processing the filter is cleaned with MilliQ water followed by rinsing it with alcohol and acetone. Punches with a diameter of 14mm were taken out, each filter provided 4 punches. The procedures for the sample filters were also performed for the filters used for the standard and blank measurements. After punching the filters, the punches were placed in an acid desiccator to remove all inorganic material and left overnight. The next day the punches are transferred to the oven and baked overnight at 50°C. The next step was the packing of the cups. For each hole in the Elemental Analyzer, two cups were prepared to increase the concentration and two punches of a filter were placed in one cup. For the calibration of the measurement an external standard was used. Seven standards were prepared for each measurement. IAEA-CH6 and IAEA-N2 was added to a cup, with a mass ranging respectively from 0.05mg to 0.6mg and 0.05 to 1.0mg. The added mass was written down for the calibration. In each cup of the standards 2 blank punches were added. During the measurement, one hole was filled with a cup with two blank punches and the added standard and a cup with only two blank punches to get similar measurement conditions as the samples.

The EuroVector, Elemental Analyzer, was used to measure the concentration and isotopic composition of particulate organic carbon and particulate nitrogen. The EuroVector EA consists of a two-reactor system, a combustion reactor and a reduction reactor respectively at 1030°C and 650°C, where all contaminants are removed and all particulate organic carbon is converted to CO2 and all particulate nitrogen to N2. The reactor columns are followed by a water trap and a gas chromatography column for the separation of the evolved gases (CO2 and N2). The thermal conductivity column measured the amount of CO2 and N2. The analysis of the carbon and nitrogen isotope ratio occurred by IRMS (Nu-Horizon) at AMGC.

Dissolved organic C (DOC)

The filtrate of the sample preparation of the particulate matter is used for the measurement of the dissolved carbon. Samples for dissolved organic carbon and δ13CDOC were shipped to Spain (University of Granada). The measurement was performed by the Andalusian Institute of Earth Sciences, according to an existing agreement with AMGC. The 40ml EPA-vials were put in the oven at 450°C for 4h and filled with the filtrate. The samples were preserved by adding 200µl of a 25% H_3PO_4 solution. DOC and δ^{13}CDOC were measured with a Thermo HiPerTOC total organic carbon analyzer, interfaced to an IRMS. DOC was converted to CO_2 with a combined UV-persulfate oxidation method in a heated reactor and the resulting CO_2 was bubbled out by a helium stream, purified in a reduction/oxidation column at 680°C, passed over a GC column and transferred to the IRMS for quantification and stable isotope measurements.

Major ion analysis

The ion analysis was carried out using an ion chromatograph (IC). Samples were melted overnight in a refrigerator prior to analysis. The samples were then analysed using a Dionex ICS5000 liquid chromatograph. The ions measured in this analysis were methane sulfonic acid (MSA), chloride (Cl^-), nitrate (NO_3^-), sulphate (SO_4^{2-}), sodium (Na^+), magnesium (Mg^{2+}) and calcium (Ca^{2+}). The cations were measured using MSA as eluent. The anions were measured using a gradient and KOH as eluent. The measurement was carried out by project partner Université Libre de Bruxelles.

3.2 Characterisation of the inorganic composition of atmospheric particles

A 250km transect was sampled from the highlands south of PEA station to the coast. Along this transect, 7 sampling sites were defined (see Table 1 and Fig. 1). Systematically, at each of these sites, a Sigma-2 passive sampler collected one-year dust deposition (on one filter/year), and snow sampling was performed (x 10L per site). At two of the passive sampling sites (Frank Kenny North and Plateau), 3-m long ice cores for chemical analyses were successfully collected. Finally, an active sampler was installed in the “atmosphere” container, around 350 m north of PEA main station, including a strong pump (nominal flow rate of 330 L/min) and an inlet on the roof of that container. About 10 filters per season were collected. All the collected samples were carefully stored in the freezers at the G-Time laboratory of Université Libre de Bruxelles (ULB, Belgium).

3.2.1 Active sampling and analysis of inorganic composition of atmospheric particles

Active sampling using 0.2 µm pore-size Teflon filters has been done in 2018, 2019, 2020, and 2021. For this active sampling, the system was improved throughout the sampling campaigns: the strong pump (nominal flow rate of 330 L/min) was connected with a slightly different inlet on the roof of the container (to guarantee the robustness of the inlet and avoiding the entrance of snow in the apparatus). A flowmeter was also installed in the sampling line in order to derive accurate values for actual and total sample flows. In addition to several blank samples, a total of 10 filter samples per season were recovered.

SEM-EDS analyses of the filters from 2018 to 2021 campaigns were performed or are planned to be performed by Sibylle Boxho at LPCA – ULCO, Université Côte d’Opale, Dunkirk, France. SEM-EDS are the abbreviations of a Field Emission Scanning Electron Microscope (i.e., SEM) (JeolTM JSM-7100F) equipped with three energy dispersive X-ray detectors (EDS; Bruker XFlash 6/30), enabling the compositional analysis of elements with an atomic number higher than boron ($Z \geq 5$), measurements of size and shape. Each spectrum was carried out with an acquisition time of 20 s, at an acceleration voltage of 15.0 kV, with a 10.0 mm working distance and a probe current of 300 pA. Calibration of EDS detectors was performed on a copper (Cu) strip fixed close to the sample filter.

3.2.2 Passive sampling and analysis of inorganic composition of atmospheric particles

The passive sampling technique for ambient-air particle collection was based on the use of Sigma-2 passive-samplers (Dietze *et al.*, 2006; Grobety *et al.*, 2010). The Sigma-2 inlets were elaborated by the Deutsche Meteorological Service (DWD; Freiburg) and were provided for this study by Volker Dietze of DWD. They guarantee collection of airborne particles in the size range of 2.5 to 80 µm.

Inside the Sigma-2 Polyvinyl Chloride cylinder, a polycarbonate filter (47 mm diameter, 0.2 µm pore size) was placed as sampling surface and fixed with teflon filter holder. Since the 2018 campaign, a savilex® beaker was also added next to the filter holder for testing another way of dust sampling. The passive samplers were installed at the top of aluminum poles at ±2.5 m height (above the snow surface), which is enough for avoiding burial of the sampling system under the snow accumulation. During the last campaigns, the poles were progressively de-installed because of snow accumulation and slightly displaced (500m) relative to the previous locations and finally removed at the end of the CHASE program.

Similarly to the active sampling filters, SEM-EDS analyses were performed on the Sigma 2 dust filters by Aubry Vanderstraeten and Stefania Gili at LPCA – ULCO. Airborne particles were systematically analysed using a combination of three analytical modes: automated single particle analysis, single particle analysis in manual mode and mapping. For reproducibility of the measurements, all automatic analyses were performed at a 2000 times magnification. The automated analysis is a fast, easy and efficient method for a first particle characterization, with chemical component measurement, particle size and shape, providing a substantial quantity of results (several hundred particles analysed per sample) requiring an efficient way to process the full dataset. Initially, clustering statistical analysis was applied to distinguish particle types and better understand the compositional variability within the whole sample sets (*e.g.*, on Antarctic dust, Dalia Pereira *et al.*, 2004). However, only mineralogical trends can be obtained this way. Therefore, to get further detailed mineralogical identification, we developed a Matlab script to classify each particle in a specific mineralogical group (quartz, aluminosilicates, Fe-bearing aluminosilicates, Fe-Mg silicates, Fe oxides, Ti oxides, metal-rich (potentially anthropogenic) groups).

In addition to the SEM-EDS semi-quantitative chemical and mineralogical composition analyses, trace element analyses were carried out by Element II ICP-MS (in collaboration with AMGC-VUB), in order to know the relative concentrations and in particular the rare earth profiles.

3.2.2 Sampling of surface snow and analysis of inorganic composition of particles therein

Surface snow samples have been collected in November - December 2017, 2018, 2020 and 2021 at 6 locations: i) around 4 km eastward of PE station; ii) one site southward of PEA, in the vicinity of the plateau; iii) 16 km from that site, a new site at the deep plateau; iv) at Romnoes; v) in between Romnoes and the coast (FKN); v) at one site at the western part of an ice rise at the coast (FKS) and, vi) 'Breid Bay', near the coastline (only in 2020 and 2021). For each of these campaigns, a total of ~30 bottles of 10 L, i.e., a total of around 300 L of surface snow, have been collected. The bottles have been shipped back, delivered and stored in our lab, where they were treated for chemical and isotopic analyses of the particles from the filtered snow samples in ultra-clean laboratory conditions. In addition, at the Frank Kenny north and the Plateau sites, 3-m firn/ice cores were drilled for the preliminary analysis of inorganic particles.

Snow samples were melted at room temperature and filtered on Whatman™ polycarbonate filters with 0.2 µm pore size and 47 mm diameter. For low amount of dust samples, the filtration was done on 25 mm diameter filters to gain a 3.5 concentration factor of particles on the total filter surface. In order to optimize the SEM-EDS individual particle analysis (amount and type) per sample filter, the latter was cut and a quarter was fixed on a metallic holder with a double-face conductive tape. These analyses allow us to compare chemical composition and mineralogy of the dust deposits with the suspended atmospheric particles (sampled by the passive and active).

However, the main analytical objective was dedicated to trace element concentration analyses, in particular rare earth element (REE) concentrations and patterns. Therefore, the rest of the filter was stored for dissolution of the dust in a (HF+HNO_3+HCl) mixture in Savillex® vials before being prepared for trace element concentration analyses (by ICP-MS Agilent 7700 at G-Time and HR-ICP-MS Element II at the VUB). A novel statistical REE model was developed to compile the REE profiles of the Antarctica dust samples with those from all related dust potential source areas from the southern hemisphere. This innovative approach helps to discriminate and quantify the PSA respective contributions to Antarctica atmospheric fallouts. It should also provide a complementary powerful tool to the isotopic tracing method for identification of the origin of the mineral dust, especially when the sample amount is a limiting factor.

In parallel, the snow samples (when larger volumes were collected) were also filtrated and dissolved for Sr, Nd and Pb isotopic measurements on a Multi Collector ICP Mass Spectrometer (MC-ICP-MS) Nu Plasma II in dry mode (DSN desolvating system). Before measurements, a new chromatographic procedure on ion-exchange resin was specifically and carefully developed for these snow samples to extract strontium (Sr), neodymium (Nd) and lead (Pb) from a single sample aliquot (to minimize the sample amount required for the isotopic analysis) (Vanderstraeten et al., 2020). In addition to the chromatographic purification methods, specific attention was paid to the low procedural blanks and the isotopic measurements themselves that require the absolutely optimal tuning conditions of the instrument and operator because of the low analyte amounts.

3.3 Source region and transport pathways analysis of atmospheric particles and VOCs

The FLEXTRA trajectory model (Stohl et al., 2005) has been applied in order to investigate possible source regions and transport pathways into Antarctica of atmospheric particles and SVOCs. The model was driven with ECMWF ERA5 meteorological fields (0.5° x 0.5° grid). 10-days backward trajectories, starting from PEA, were calculated for the period 01/01/2010 to 31/12/2020, in 6-hour-intervals. A k-means cluster analysis has been done based on several parameters, following the method in Delcloo and De Backer (2008). The cluster analysis has been done for the whole period (2010 – 2020) and also for each season separately. All 10 days of the back trajectory calculation have been used for the clustering. When relying on the normalised latitude, longitude and altitude, four clusters of air mass origin have been found. In addition, the backward trajectories have been combined with distinct parameters like particle number concentration, aerosol absorption exponent, potential vorticity or exposure to sunshine duration.

Further, the FLEXPART dispersion model (Stohl et al, 2005) has been applied in order to identify potential source regions, e.g., for the individual sample periods of the active sampling during seasons 2017/18 up to 2020/21. FLEXPART is a Lagrangian particle dispersion transport model which is originally designed for calculating the long-range and mesoscale dispersion of air pollutants from point sources. FLEXPART was run 30 days back in time starting from the end of each measurement/sample period. Wet or dry deposition was not taken into account and the model was run with passive tracers. In these cases the dispersion analysis yielded probability functions for the source regions and atmospheric pathways.

3.4 Further instrumentation at Princess Elisabeth Station

Physical properties of atmospheric aerosol particles have been measured at PEA since 2010 by partner RMI. Measured properties include total particle number (3 – 3000 nm size range; Condensation Particle Counter, TSI3776), particle number size distribution (90 – 7000 nm size range, Laser Aerosol Spectrometer, TSI3340), and the aerosol light absorption and scattering coefficient (Magee AE-31 aethalometer and Ecotech Aurora 3000 nephelometer, respectively). Measured atmospheric aerosol properties have been further analysed in order to link them to the number of cloud condensation nuclei, based on the paper by Herenz et al (2019).

In addition, filter samples have been collected for the laboratory analysis on ice nuclei particle (INP) concentrations. The collected samples have been analysed by TROPOS, Leibniz Institute for tropospheric research, Leipzig, Germany, thanks to collaboration between them and RMI. First samples were taken during field season 2017/18. Based on the made experience, sample time (10 days) and set up (pump, flow, and filter treatment) have been adjusted for field seasons 2018/19 and 2020/21. A total of 12 samples could be collected.

These aerosol measurements were complemented by data of an automatic weather station (sub-contracted by RMI to Utrecht University, Institute for Marine and Atmospheric Research Utrecht, Netherlands). Also, vertical profile measurements of temperature, humidity, wind and pressure have been performed during austral summer seasons by radio soundings with weather balloons at PEA.

4. SCIENTIFIC RESULTS AND RECOMMENDATIONS

4.1 Characterisation of the organic atmospheric composition (particulate matter and VOCs)

4.1.1 Active sampling of semi-volatile organic compounds (SVOCs)

A total of 32 PUF and 32 QFF samples were collected using the HVS during the summer-only measurement campaigns. Each sample represents the gas or particle phase over 1 week. All samples were analysed twice on the GC-HRMS using a separate method to quantify PAHs and oxy-PAHs. Great care was taken to ensure low background and detection limits and high analyte recovery from the sample media. The limit of detection (LOD) was well below 1 pg/m³ and recoveries ranged between 70 and 100% for all compounds except naphthalene. 22 compounds were quantified in all PUFs (10 PAHs + 12 OxyPAHs) which makes this the largest dataset on PAHs and related compounds in Antarctica.

Table 2: overview of amount of filter samples taken via active sampling during the individual field expeditions

Sample season	# Samples + Blanks
2017-2018	9 + 2
2018-2019	6 + 2
2019-2020	9 + 2
2020-2021	8 + 2

A general conclusion is that the amount of PAHs and oxy-PAHs in the particle phase was found to be negligible. This was probably due to the very low particle number concentration in the air around the station as shown by earlier measurements (Herenz et al., 2019). Fluorene, phenanthrene, fluoranthene and pyrene are the most ubiquitous compounds found in the samples with concentrations ranging between 1 and over 100 pg/m³ (see Figure 2). No significant inter-annual differences are found for these compounds. Examining the link between the meteorological conditions during the time of sampling and the measured concentrations is a challenging task requiring more sampling.

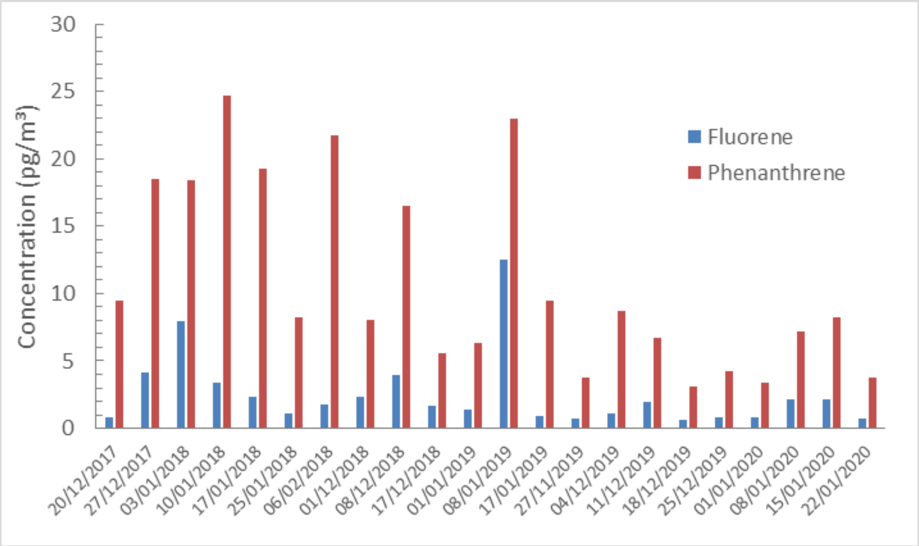

Figure 2: concentrations of Flourene and Phenantrene for the austral summer seasons 2017/18, 2018/19 and 2019/20

4.1.2 Passive sampling of semi-volatile organic compounds (SVOCs)

During method development it became clear that high extraction efficiencies were difficult to attain for the PDMS sheets using PLE. Higher temperatures and stronger solvent combinations degraded the sample medium resulting in a high amount of matrix material in the extract. Poor contact of the solvent with the PDMS sheet was probably the root cause of the poor analyte recovery. A metal grid was inserted in the ASE cells resulting in a slight increase in extraction efficiency.

The PUF foam filters were all collected and stored in a freezer until analysis. As the reported idealized sample rate for a PUF filter is 4m^3/day we extracted the two filters of one site in one cell to obtain a similar equivalent sample volume compared to the HVS. The same extraction methodology was validated using reference material. This showed very good analyte recovery. Upon collection it was noted that the polyurethane was severely discoloured from white to a shade of yellow. During sample extraction a large amount of non-volatile matrix material was co-extracted which wasn't present in blanks nor in the extracts of earlier field tests. Artificial aging of the PUF for several weeks with a UV-light showed similar yellowing and matrix co-extraction, indicating the degradation of the PUF filters exposed in Antarctica is caused by indirect UV radiation. A clean-up method was developed by evaluating different adsorbent cartridges (Florisil and EZ-POP NP) and different eluent compositions. Recovery of PAHs and oxy-PAHs was measured based on the addition of reference material to the cartridge. Finally, the method was validated both with laboratory surrogate samples (cleaned PUF filters which were exposed for 2 weeks to a UV light source) and in-situ surrogate samples (conditioned PUFs exposed in Antarctica for 1 week without shelter). During the extraction and volume reduction of actual samples exposed for a year no co-elution was observed after clean-up. However, when analysed on GC-HRMS, some unidentified compounds interfered with the measurement. Further method development is required to properly analyse these samples, an alternative analytical instrument such as a GCxGC separation or a faster HR-MS (ideally a combination of both) would also likely resolve the interference.

4.1.3 Passive sampling of volatile organic compounds (VOCs)

4 sets of samples consisting of axial sorbent tubes and blanks were collected over the period of the project. Before analysis samples were dry-purged with a validated method to remove any traces of water which might have gotten stuck in the tube or sorbent during exposure or sample collection. The dry-purge proved highly successful with not a single sample rejected because of water. On each passive sample site at least 6 samples + 2 blanks were taken. 3 samples and 1 blank per site were analysed on TD-GC-MS while the other 3 samples and blanks were analysed on the novel TD-PTR-TOFMS. The results for both instruments show high similarities except for in some cases where detection with the more sensitive PTR-TOFMS yielded data which was below LOD for the TD-GC-MS. On the other hand, the chromatographic separation of a GC allows for the distinction between various isomers present in the sample. PTR-TOFMS is based on the proton transfer reaction, a consequence is that halocarbons and alkanes could not be detected using this technique. Overall TD-PTR-TOFMS proved to be a very complementary technique to TD-GC-MS.

Table 3: overview of sites from where passive samples were collected and for the different field expeditions to Princess Elisabeth station; see also table 1; BreidB=Breid Bay; FK.North=Frank Kenny North; FK.South=Frank Kenny South; Romn=Romnoes; PEA=Princess Elisabeth station; PLA=Plateau; D.Pla=Deep Plateau; CLIMB=site for remote sample collection for the Belspo Brain-be project CLIMB

	BreidB.	**FK.North**	**FK.South**	**Romn.**	**PEA**	**PLA.** †	**CLIMB**	**D.PLA**†
2017-18		X	X	X	X	X		
2018-19	X	X	X	X	X	X		X
2019-20		X	X	X	X	X		X
2020-21		X			X		X	X

Over the first 3 years 158 samples were collected in which around 65 compounds are identified and if possible quantified. This results in a dataset of over 10000 data points making it the largest dataset on VOCs and oxy-VOCs in Antarctica by some margin.

In general, it can be concluded that oxygenated aromatic compounds are by far the most important group by concentration. Acetophenone, phenol, benzaldehyde and benzoic acid are known oxidation products of primary aromatic compounds and are present in concentrations up to 2 µg/m³. Additional data analysis is required to discover if significant spatial or inter-annual trends are present. For example, dimethylsulfone ($DMSO_2$), an oxidation product of dimethylsulfide (DMS), clearly shows a decreasing trend in function of the distance of the sample site to the ocean (Fig. 3). A good separation between the sample sites under coastal and continental influence is shown when using a dimension reduction technique such as principal component analysis (PCA) on the data (Fig. 4). Recent advances in the processing of the MS data generated by PTR-MS (which contains several orders of magnitude more data compared to GC-MS) enables a deeper understanding of the data.

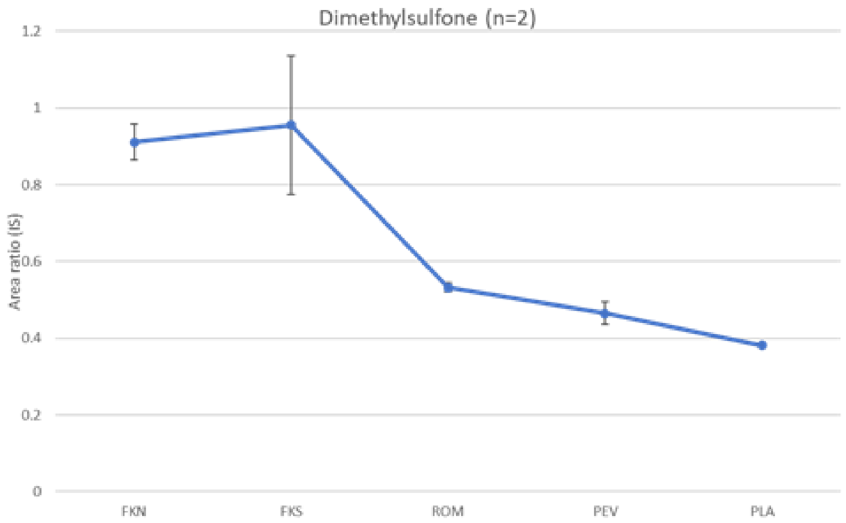

Figure 3: amount of dimethylsulfone ($DMSO_2$), an oxidation product of dimethylsulfide (DMS), in function of the distance of the sample site to the ocean; FKN=Frank Kenny North; FKS=Frank Kenny South; ROM=Romnoes; PEV=Princess Elisabeth station; PLA=Plateau (see also table 1)

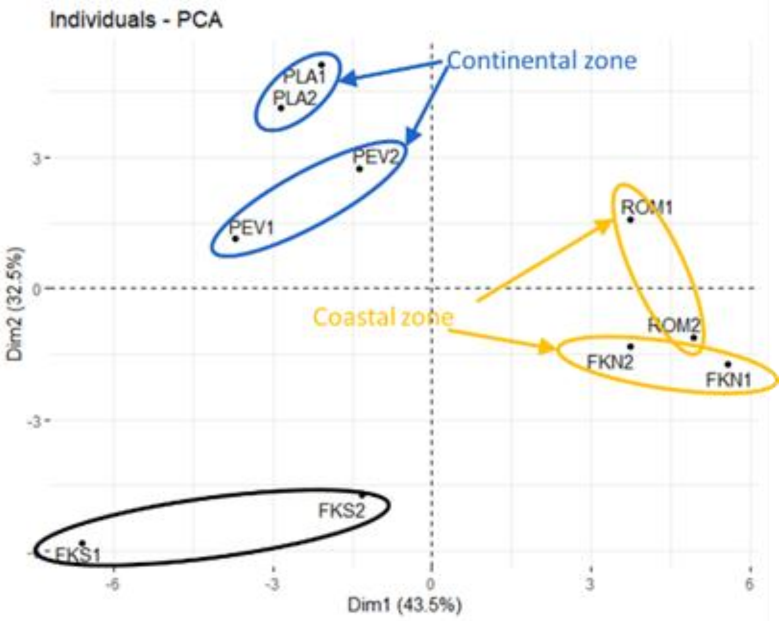

Figure 4: separation of the different passive sampling sites by principal component analysis

4.1.4 Concentration, distribution and sources of organic carbon in East Antarctic snow and stable isotopes C and N of organic fraction

The sampling points from the Donning Maud Land (DML) ranged from 5km to 230km from the sea. POC concentration in surface snow samples ranged from 3.1 ± 0.5 to 22.8 ± 0.3 µmol l-1 with a mean concentration of 9.79 µmol l-1 (117.46 µg l-1). DOC concentrations in surface snow samples from the same transect ranged from 23.7 ± 0.5 to 61.9 ± 0.5 µmol l-1 with a mean concentration of 32.8 µmol l-1 (393.7 µg l-1). The δ13C is given for both POC and DOC with values respectively ranging from -27.24 ± 0.88 to -22.34 ± 4.51‰ and -27.97 ± 0.1 to -23.64 ± 0.03‰. The analysis of 2017 and 2019 samples gave similar results, except for one outlier with the δ13C DOC. The particulate nitrogen concentration ranged from 0.29 ± 0.23 to 1.09 ± 0.49 µmol l-1.

Most ions, Na+, Cl-, K+, SO4^{2-}, decrease with increasing distance from the sea. The right panel shows the ion concentrations of 2017. The concentration is similar as the concentration of 2019 but no decrease with increasing distance is observed. Na+ is considered to be the most conservative ionic proxy for sea spray in coastal Antarctica so this will be used to determine the sea spray contribution to the organic matter. The mean annual accumulation rate is used to calculate the fluxes for POC, DOC and ssNa+. Both the POC-flux as the DOC-flux decreases with increasing distance from the sea and increasing altitude (Figure 5). The DOC-flux has a higher correlation with both distance from the sea and the altitude compared to the POC-flux.

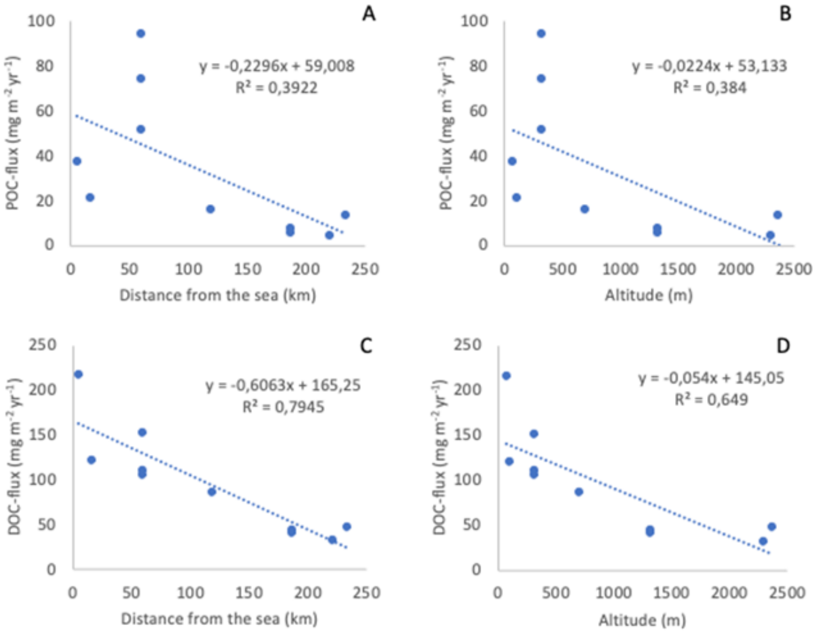

Figure 5: The correlation of the POC-flux with the distance from the sea (A) and the altitude (B) and the correlation of the DOC- flux with the distance from the sea (C) and the altitude (D)

The POC and DOC concentrations show no correlation with the distance from the sea. When flux is plotted as a function of distance from the sea, a linear correlation is observed for DOC. This suggests the sea as a possible source for DOC. The POC-fluxes in function of the distance from the sea varies greatly with a general decrease trend.

Deposition of sea-spray aerosol is often inversely correlated with the distance from the sea and the altitude. The sea-salt contribution of Na$^+$, ssNa$^+$, calculated for the CHASE snow samples shows the same trend as a function of the distance from the sea or the altitude (Figure 6). There is a lot of variation in the plot that result in a low R^2 value for both the ssNa$^+$ concentration and the ssNa$^+$ flux but a general decrease is observed. The ssNa$^+$ fraction of the total Na$^+$ was found to be >70% for most sites, which indicates that a substantial portion of Na$^+$ in these snow samples is derived from sea-spray. Only the deep plateau sample, from the sampling point located the furthest from the sea, gave a $ssNa^+$ fraction below the 70% cutoff.

The correlation between $ssNa^+$-flux and both the POC-flux and the DOC-flux is plotted in Figure 7. The linear correlation between $ssNa^+$-flux and DOC-flux is higher (R^2=0.7037) than the linear correlation between $ssNa^+$-flux and POC-flux (R^2=0.1232). The high correlation between the $ssNa^+$-flux and the DOC-flux suggests that sea spray aerosol is the main source for DOC present in the surface snow samples. This also suggests that DOC and $ssNa^+$ are simultaneously removed from the atmosphere. A possible explanation for the low correlation between POC and ssNa+ can be found in the particle sizes. Both DOC and ssNa+ are mainly in the super-micron size range (Facchini et al., 2008), which leads to comparable atmospheric lifetimes. The POC particles are mainly in the submicron size range, so they are transported over longer distances in the atmosphere (Seinfeld and Pandis, 2016).

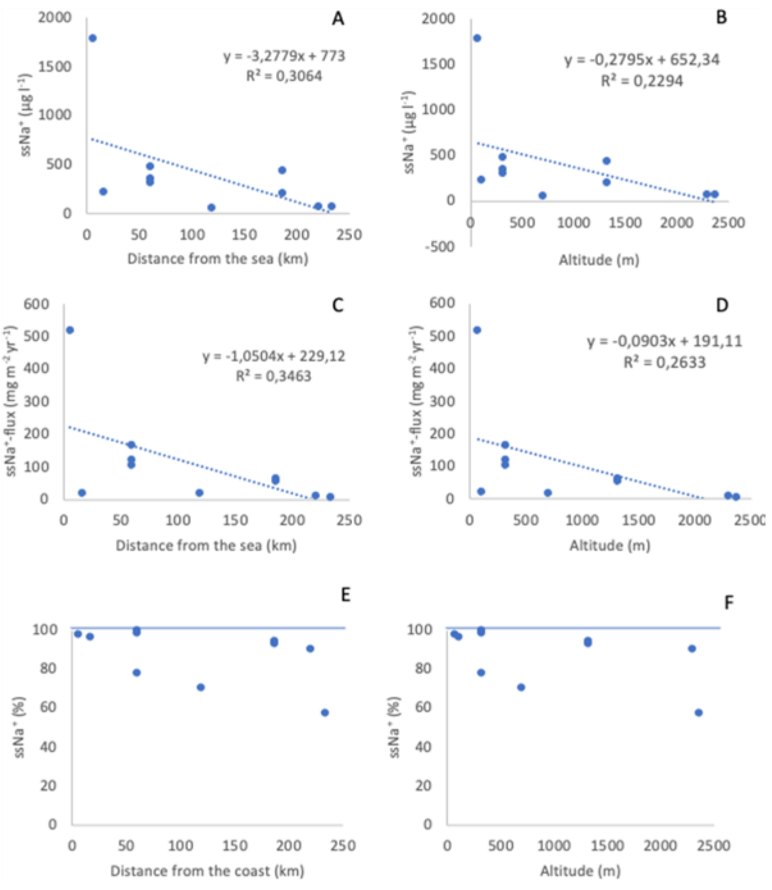

Figure 6: The $ssNa^+$ (A), $ssNa^+$-flux (C) and $ssNa^+$ - ratio (E) in function of the distance from the sea. The $ssNa^+$ (B), $ssNa^+$ -flux (D) and $ssNa^+$ - ratio (F) in function of the altitude.

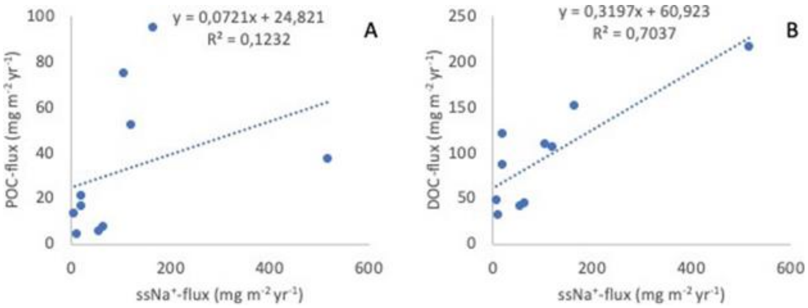

Figure 7: The POC-flux (A) and the DOC-flux (B) in function of the ssNa$^+$ -flux

The wet and dry contribution to the net depositional fluxes can be estimated. Figure 8 shows the correlation between the total flux and the accumulation rate. An exponential function represents the trend of the snow samples and provides the contribution of dry deposition to the total flux. The fraction dry deposition of the mean total flux was 6.8% and 23.3% for respectively POC and DOC. The numerical value of this fraction is not accurate because simplifications were made to obtain the function but the general observations are probably valid. The contribution of dry deposition is minor compared to the contribution of wet deposition. The dry deposition for DOC is higher than for POC because DOC is in the super-micron size range, so more susceptible for gravitational settling (dry deposition, see Seinfeld and Pandis, 2016)

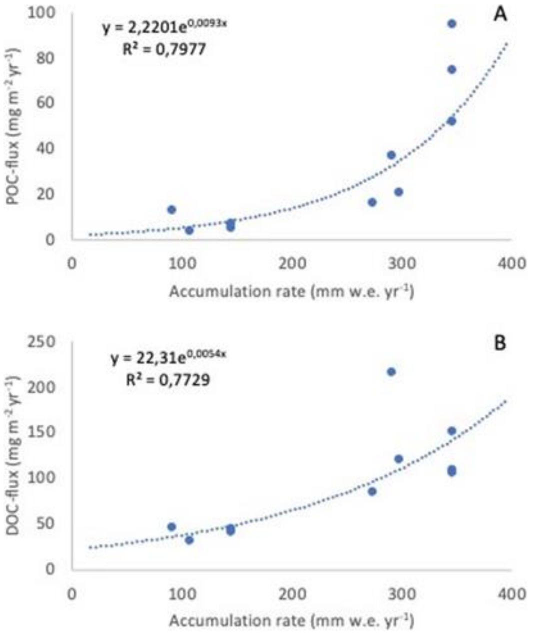

Figure 8: The correlation of the POC-flux (A) and the DOC-flux (B) with the accumulation rate

The particulate nitrogen concentration was too low, so it was unreliable to be used for spatial distribution or source identification. It still can be a useful tool to determine the C/N ratio for identifying carbon sources (Figure 9).

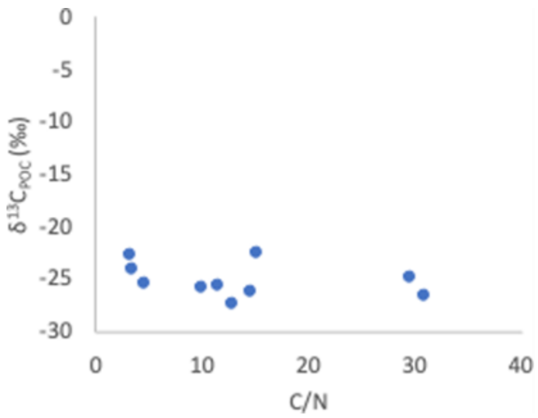

Figure 9: Correlation between δ^{13}C-POC and C/N

Organic carbon has a lot of possible sources and is involved in different bio-geo-chemical processes before and after deposition in the snow. The isotopic composition can be a useful tool to try to identify the source. Figure 10 shows the correlation between δ^{13}C-POC and the natural logarithm of the concentration of POC. The plot shows a significant correlation (R^2=0.6985). This means that POC has undergone fractionation processes. The enrichment factor for POC in snow is -1.93‰. A possible process causing this fractionation is the oxidation of VOCs. This process leads to a depletion of the formed organic compounds (Fisseha et al., 2009; Iannone et al., 2003). The magnitude of the depletion depends on the molecule and the oxidation reaction.

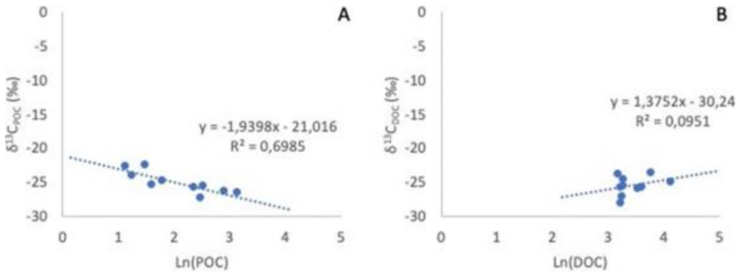

Figure 10: (A) The correlation between $\delta^{13}C_{POC}$ and Ln(POC concentration). (B) The correlation between $\delta^{13}C0_{DOC}$ and Ln(DOC concentration)

The POC originating from the ocean has a $\delta^{13}C_{POC}$ value of -22 ‰. The $\delta^{13}C_{POC}$ of the snow samples ranges from -27.24 to -22.34 ‰. The correlation between POC-flux and ssNa$^+$-flux was low, so other sources probably have a significant influence on the POC present in the snow, such as input of organic carbon by local microbial activity and by long-range atmospheric transport of organic carbon from other continents (Antony et al., 2014). Further investigations will be needed to link isotopic ratios to these possible sources. Figure 10 plotted $\delta^{13}C_{POC}$ in function of the C/N ratio. Figure 10B shows the correlation between $\delta^{13}C0_{DOC}$ and the natural logarithm of the concentration of DOC, but no correlation is observed. This suggests that it is not influenced by fractionation processes. Primary sources are less depleted in ^{13}C due to the absence of significant fractionation (Fisseha et al., 2009), indicating that the DOC is mainly originating from primary aerosol. The DOC has a similar isotope ratio as its source. From the correlation between the DOC-flux and ssNa$^+$-flux, sea-spray was identified as a main source for DOC in snow. Plankton emits marine primary organic aerosols, which can be a major source for water soluble organic carbon, which can explain DOC present in the surface snow samples. The carbon isotopic ratio for organic aerosol emitted by plankton is -31 to -18 ‰. The $\delta^{13}C$ for DOC ranges from -27.97 to -23.64 ‰, which is completely within the range of the plankton emissions.

4.1.5 Conclusions and Recommendations

Volatile and semi-volatile organic compounds

The measurement of PAHs and oxy-PAHs in an extreme environment such as Antarctica proved to be very feasible. It would be useful to further extend the dataset to get a better correlation between the back-trajectories an observed PAH and oxy-PAH concentrations. The high-volume sampler proved to be able to operate without extensive maintenance for at least 4 years. Only two minor repairs were necessary. Long time series and big data are an important driver in modern climate science.

We demonstrated VOCs and breakdown products do occur in Antarctica but to better understand particle formation and the interaction with ice condensation nuclei and cloud formation and cloud microphysics a time series with a higher temporal resolution (days to a week) is required. Ideally such a measurement would take place both at the coast as well as on the plateau at the same time as we proved these areas are distinctly different. Measurements like these, using high-end analytical equipment and innovative sampling methodologies help to unravel the complex atmospheric chemistry in remote areas.

Concentration, distribution and sources of organic carbon in East Antarctic snow

This study remained preliminary, mainly due to the limited amount of available samples. Nevertheless, it showed the significant influence sea spray on the POC and DOC present in surface snow. Especially the linear correlation between the DOC- flux and ssNa+-flux indicates that sea spray was the main source of DOC. This is confirmed by the carbon isotopic ratio of the DOC. The absence of fractionation suggests that primary organic aerosol, originating from plankton emission, was the main contributor of DOC.

No clear source was identified for POC. The analysis showed that POC was influenced by both marine and continental sources. The carbon isotope ratio showed a depletion in ^{13}C, indicating that the ratio was affected by fractionation processes.

This study primarily focused on evaluating the contribution of marine aerosol, but literature suggests also the presence of other significant sources of organic carbon that might affect the POC and DOC. It is important to remember that the organic carbon concentration and the isotope ratio are also subjected to post-depositional processes. This study provided carbon isotope ratios of POC and DOC in surface snow samples for the first time in the region around the PE Station. It can act as a starting point for further investigation of all processes involving POC and DOC in Dronning Maud region of East Antarctica. Clearly a compound specific analysis is likely to shed more light on the question. However, this would require a set of new, larger samples of snow, taken for the specific purpose to carry out stable isotopes analyses.

4.2 Characterisation of the inorganic composition of atmospheric particles

4.2.1 SEM-EDS analyses: size, shape and mineralogy of atmospheric particles (in suspension or deposition) collected by passive and active samplers

The large majority, up to 89 %, of the particles are below 2 µm and up to 50 % of particles are of submicron size. Practically no particle with a size > 5 µm was detected using automatic analyses. As a whole, the particle size fractions along the 250 km transect show no significant distinction and can be considered as similar (Figs. 11 and 12). The same observation is made when comparing passive samplers to snow filtration at the exception of higher amount of 0.25-0.49 µm to 0.5-0.99 µm particles for passive samples compare to snow samples.

The large majority of atmospheric particles are characterized by angular shapes and faces. A low proportion of spherical particles was also reported in most samples and corresponded systematically to Ti oxide nanoparticles. Some diatoms or pieces of them were observed.

The automated spectral acquisition favours the risk to mix up chemical compositions of single particles within an aggregate. On one hand, the overlapping of particles could not change the mineralogical classification, especially when the major cation was present in both particles. As an example, the juxtaposition of additional Fe oxides did not change the mineralogical group identification as Fe belongs also to the crystalline structure of the bigger particle (illite). But, as a consequence, the occurrence of small iron oxides would be underestimated. On the other hand, when the chemical compositions of two agglomerated particles are distinct, the final result can lead to an erroneous mineralogical identification. However, among all particles analysed clear mineralogical groups are revealed: aluminosilicate, silica, titanium and iron oxides particles are identified as “pure” particles. However, even though all these particles are considered as pure particles, they often contain low fractions of unexpected elements, like iron, aluminum and silicon. As stated by Deboudt et al. (2010), no really pure aerosols exist in the atmosphere because internal mixtures are almost systematically present.

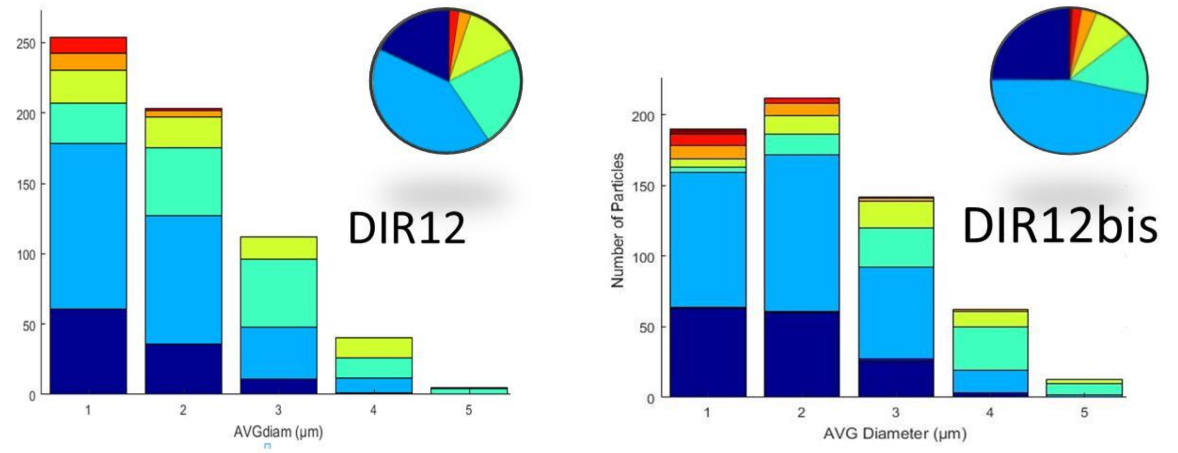

Figure 11: Histogram of size fraction and chemical classification of DIR12 (passive sampler) and DIR12bis (snow samples). Color legend is available in Figure 12. The size groups based on the average diameter are defined as follow: 1) 0.25 - 0.49 µm, 2) 0.5 - 0.99 µm, 3) 1 - 1.99µm, 4) 2 - 3.99 µm, 5) ≥ 4 µm.

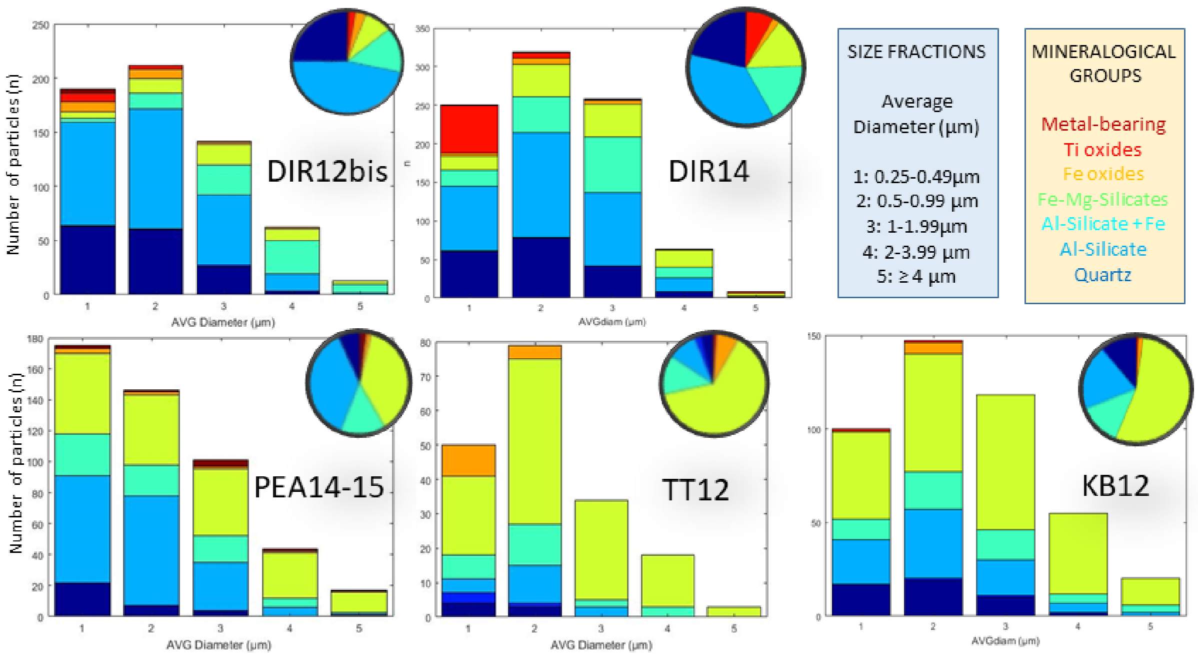

Figure 12: Size fractions (x-axis) and mineralogical group (color) distribution of particles (n in y-axis) analysed in automatic mode

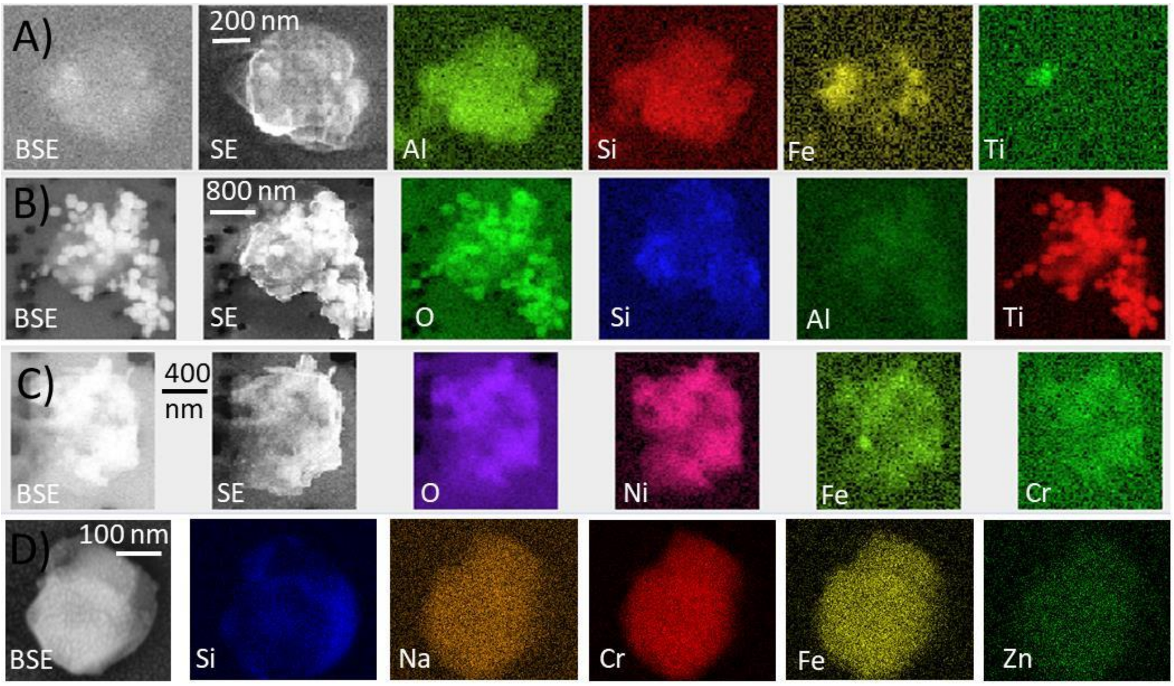

Figure 13: Metal bearing particles presented as a Mosaic of mapping images. BSE and SE stand respectively for backscatter electron and secondary electron.

However, the entire sample set presents a comparable mineralogical composition dominated by aluminosilicate, silica and Mg-Fe silicates closely followed by Fe-bearing aluminosilicates and iron or titanium oxides. In a much lesser proportion and non-systematically, metal-bearing particles composed of Cr, Ni, Zn, Cu, Sb, Sn, Tl, Ta are present and supposed to have an anthropogenic origin. Also, comparison between the SEM-EDS results obtained from filters of the Sigma 2 collectors and snow samples display no significant size, or chemical composition difference (Figure 13).
Special attention was drawn to particles containing Fe as iron is indeed a key micronutrient essential for the primary productivity in the Austral ocean, the largest High Nutrient Low Chlorophyll area (HNLC) (Martinez- Garcia *et al.*, 2014). In addition to the large occurrence of iron in the crystalline structure of silicates and iron oxides, iron coating on particles was also observed. Overall, it is clear that the occurrence of Fe-bearing particles is widespread in East Antarctica, mostly as internal element in the crystalline structure of Fe-aluminosilicates or Fe-Mg silicates, also as iron oxides (Fig. 13) and finally in the form of Fe-coating.

A key observation concerns the mineralogical differences between samples originating from the coastline and those from inland. Coastal samples show a predominance of aluminosilicates and quartz (> 75 %) accompanied by a weaker proportion of Fe-Mg silicates (< 20 %) as reported in Figure 11. In contrast, inland samples exhibit a strong enrichment in ferro-magnesian aluminosilicates (Fe-Mg silicates > 50 % - Fig. 12), which clearly exceed the proportion of Fe-bearing aluminosilicates, aluminosilicates and quartz. This observation is consistent with the Sør Rondane mountains at the vicinity of these sampling sites, which display a large variety of lithologies with a main occurrence of amphibolites and gneisses mainly composed of melanocratic Fe-Mg rich minerals (Satish-Kumar et al., 2008). Typical long-range transport dust is generally composed of feldspar (fsp), quartz (qtz), clay minerals, carbonates, sulphates, phosphates, salts and also amphiboles (am) and pyroxenes (px) (Claquin *et al.*, 1999; Formenti *et al.*, 2014). The predominance of feldspar and quartz observed for coastal samples and a low proportion of Fe-Mg silicates (< 20 %) (Fig. 11) is in favor of a distal origin. For comparison, at Epica Dome C, the average mineralogical compositions are made of clays (illite (32 %), chlorite (5 %), kaolinite (1.5 %) and smectite (1.3 %)), feldspar (18 %), quartz (13 %), px-am (3 %) and metallic oxides (2.5 %) (on n = 225 - Gaudichet *et al.*, 1986).

The long-range transport feature of the particles is also suggested by the dominant fine size fraction and the angular shape of the majority of the examined particles. Submicron size fraction and angular shape were typical features previously reported for aerosols transported on long distance (Mahowald *et al.*, 2014; Maring, 2003). These results are in agreement with observations reported at the Epica dome-C (Delmonte *et al.*, 2004). Surprisingly, even samples collected at the three inland sites, very close to the Sør Rondane mountains, show more than 50 % of submicron-sized particles. Schwamborn *et al.* (2012) suggested that a process of repeated strong freeze-defrost cycles enhances the rock denudation, especially in mountainous environment, and can generate fine particles, which could explain this particular characteristic of the inland samples.

Finally, compared to the spatial variations displayed along the transect in terms of sample mineralogy, the temporal variations examined throughout the samples collected in 2015 through 2020 show no major fluctuations in size, shape or mineralogy of the dust deposits.

4.2.2 Geochemical analyses of snow dust samples: development of a novel statistical model based on the rare earth element (REE) patterns

Snow samples collected at the passive sampling sites along a 250 km transect from the coastline to the inland Sør Rondane mountains in NE-Antarctica were analysed for REE composition. This transect covers the transition between cyclonic (coastal) and anti-cyclonic (interior) weather systems. So far, most of the efforts to trace dust from ice cores have been focused on the central eastern regions of Antarctica, while the provenance of dust reaching the NE Antarctica coast remains poorly known although it probably better reflects the atmospheric fallouts into this sector of the Southern Ocean (i.e., Riiser-Larsen Sea).

The Sør Rondane mountains are located parallel to the coastline around 200 km inland and are mainly exposed to katabatic winds from central Antarctica (Gorodetskaya et al., 2013), making the Sør Rondane mountains an important source of local dust. Around ten Antarctic rocks and two cryoconite samples were collected and analysed for their REE patterns (and isotopic compositions) to provide a geochemical signature of the local environment, to be distinguished from distal sources. Near the Antarctic coastline, the atmospheric circulation is dominated by synoptic winds, which are able to transport dust from distal sources across the southern hemisphere (Asmi et al., 2018). Over the last few decades, it has been demonstrated the main distal dust contributor to the Southern Ocean and Antarctica is mainly southern South America (SSA) (e.g., Gili et al., 2017, 2016; Gaiero, 2007; Delmonte et al., 2019 and references therein; Vallelonga et al., 2010). Secondary contributions from other Potential Source Areas (PSAs) were also suggested, such as Australia (AUS), New Zealand

(NZ), Southern Africa (SAF) and a local Antarctic source (e.g., Basile et al., 1997; Delmonte et al., 2004a; Revel-Rolland et al., 2006). Revisiting the literature, it became apparent that there is still no consensus, which of these PSAs contribute to dust deposition over Antarctica -beyond the SSA source- mainly due to the lack of systematic studies of specific PSAs and the very small quantity of dust that is actually deposited over Antarctica.

Therefore, we established a large database by the compilation of REE patterns of all PSAs reported in the literature. In addition, we analysed the REE signature of 18 sediment samples collected in three coastal basins of Namibia, SAF (Huab, Omaruru and Kuiseb), which were recently identified as major dust source regions to South Atlantic Ocean and possibly Southern Ocean (von Holdt et al., 2017). Sediments of those three basins were re-suspended in the CESAM dust chamber (Université Paris Cité and Univ Paris Est Creteil) from which the airborne fraction was collected for analysis. In addition, we proposed a novel statistical approach compiling the REE patterns from 31 PSAs that are statistically evaluated against about 20 modern dust samples recovered along a 250 km transect in North-East Antarctica, from the Princess Elisabeth station to the coastline. Correlation coefficient (R), Akaike and Bayesian Information Criterion (AIC-BIC) were used to determine whether a dust sample traces a single or multiple PSAs. Our analyses indicated that local sources control the dust in the inland section of the sample locations, while distal sources dominate the coastal sites. Distal source areas include the previously identified Puna Altiplano, Patagonia but also Southern Africa, which is identified for the first time in recent dust in NE-Antarctica.

Based on this innovative and promising approach, we revisited the dust REE dataset of Epica Dronning Maud Land (EDML) and Epica Dome C (EDC) ice cores reported in Gabrielli *et al.* (2010) and Wegner *et al.* (2012) in order to construct the first continuous, quantitative record of dust provenance in East Antarctica during Last Glacial-Interglacial Transition (7.5-26.5 kyr) on the basis of the REE patterns. Those previous studies could not decipher the provenance of dust found in EDML and EDC ice cores from their dust REE patterns alone or REE ratios. We assembled a large database of 207 REE patterns (as Dust Source; DS) spanning across well-known PSAs of the Southern Hemisphere. Mineral dust or fine fraction (<5 µm) of loess, sediment or soils were prioritized. The fitting of dust REE pattern can be defined as a constrained least squares problem where the coefficients are not allowed to become negative (*lsqnonneg* function from Matlab). To assess the impact of uncertainties (from the REE concentration analyses) on the results of our model, we performed Monte Carlo (MC) simulations for each dust sample. To evaluate the goodness of the selected fit to a given DIC vector (the 14 REE concentrations of dust ice core), we calculated the correlation coefficient (R) at each MC iteration. Cumulated over 5000 MC iterations (per dust sample), those R values form a distribution from which we use the median R value to evaluate the overall goodness of the selected fits proposed by our model. We reveal that dust composition was relatively uniform throughout the LGM and Heinrich Stadial 1 (HS1) and strongly dominated by high-latitude PSA - i.e., Patagonia (and to a much lesser extent New Zealand), with the remaining being supplied from Australia, Southern Africa and Puna-Altiplano. The most striking change in dust provenance occurred after HS1 around 14.5 kyr BP when Patagonia and New Zealand dropped abruptly throughout the second half of the glacial-interglacial transition and into the Holocene while low-latitude PSA (Southern Africa, Australia and Puna-Altiplano) increased their contributions. Thus, besides Patagonia that remained the most important PSA, Southern Africa closely followed by Australia appear as important secondary dust sources to Antarctica.

4.2.3 Isotopic analyses: identification of additional major Antarctic dust Potential Source Areas

As previously explained, there are still many unknowns about the origin and long-distance transport of the dust that reached Antarctica. This can be explained by the extreme conditions encountered during the sampling campaigns and also by the extremely low concentrations of trace elements in snow/ice dust samples, especially during interglacial periods and consequently for modern samples. Such studies are real challenges in the field and in the laboratory. Fortunately, we were able to sample more than 100L for at least two sites per season, which give us the opportunity to develop isotopic applications providing a new Sr, Nd and Pb isotope dataset from Antarctic dust samples, but also from dust PSAs.

Revisiting the literature, it became apparent that while most of the studies have been focused on the detailed study of SSA, AUS and most recently NZ dust sources (e.g., Gili et al., 2016, 2017; De Deckker et al., 2010, De Deckker 2019, 2020; Koffman et al., 2021), other PSAs, such as Southern Africa (SAF), have been overlooked. Thus, there arises a compelling need for more extended geochemical studies to fill the current gap in understanding the past and present long-range transportation of mineral dust particles to the high-latitude environments. For many years, to investigate the role of SAF as dust PSA, only the Makgadikgadi Pan in Botswana and the Etosha Pan in Namibia were recognized as persistent dust sources in SAF (e.g., Prospero et al., 2002; Bryan et al., 2003; Mahowald et al., 2003; Li et al., 2008), neglecting the coastal Namibian sources. In fact, it was only until recently that the Namib Desert coast has been identified as one of the largest sources of dust in Southern Africa (Vickery et al., 2013; Von Holdt et al., 2017). Using remote sensing satellite images, these authors have found that along the south-western coast of SAF, the Kuiseb, Omaruru, Huab Rivers beds and Namib Sand Sea are the main areas of greatest dust emission located on recently deposited fluvial surfaces.

Through a precious collaboration with University of Montreal (Prof. J. S. King), riverbed sediments were collected along these major dust-producing river systems in the Namib desert (Huab, Omaruru, Kuiseb and Namib Sand Sea). Combining rare earth elements (REE) concentrations and the Sr-Nd-Pb isotope compositions, we seek to evaluate the influence of this region as a dust source to the high latitude environments of the Southern Hemisphere. We also include a re-analysis of the isotopic composition of SAF samples previously reported in Delmonte et al. (2004a) and Vallelonga et al. (2010). Based on these results, we compared the geochemical signature of bulk and re-suspended aerosol fractions from SAF, with the dust fingerprint reported for Antarctic ice cores (Vostok and EDC) during glacial (MIS2-MIS20) and interglacial periods (Holocene and MIS 5.5). In addition, we compared the SAF and other PSA isotopic signatures, with the results obtained on our collection of modern snow samples. These unique results were obtained thanks to a combination of valuable collaborations that ensured access to these SAF samples, successful field missions in Antarctica, and analytical procedures specifically developed for these precious Antarctic dust samples within the G-Time laboratory (Vanderstraeten et al., 2020).

The new Pb isotope data of SAF samples (Gili et al., 2022) define a field that, together with Southern South America PSAs can better explain the entire range of Pb isotopic compositions observed in the East Antarctic dust from Vostok and EDC (Figure XXX). In detail, our results show that East Antarctic dust samples show two main compositional trends that pinpoint different end-members. This is, a common end-member with low values in both 208Pb/207Pb, and 206Pb/207Pb elucidated by either the Namib Sand Sea or the Patagonia/Southern Central Western Argentina sources in combination with a more radiogenic end-member, that is, either Kuiseb PSA or the local Antarctic volcanism with a contribution from the Puna Altiplano Plateau (PAP) and the Huab region, especially during interglacial times (Figure 14). The strontium and Nd isotopic data confirm and refine these trends. In general, during the last glacial climatic periods of the late Quaternary, East Antarctic data is very uniform converging to an unequivocal signature from Patagonia with additional inputs from other lower latitude areas in SSA, such as, the Southern Central Western Argentina (SCWA) and Southern Puna (Fig. 14). On the other hand, the East Antarctic interglacial dust fingerprint, as well as the glacial/interglacial signature of marine sediments from the South Atlantic, are marked by a variable mix of sources (Gili et al., 2022). A tri-mixing hyperbola with two common end-members can be drawn and help to decipher this variability. The first mixing component is a mafic-like end-member represented by Patagonia and/or SCWA, in addition to a second crustal-like end- member, defined by the Southern Altiplano, plus a third end-member, which can either be the Northern Puna and/or Southern Africa region as a whole (dotted black line; Fig. 14).

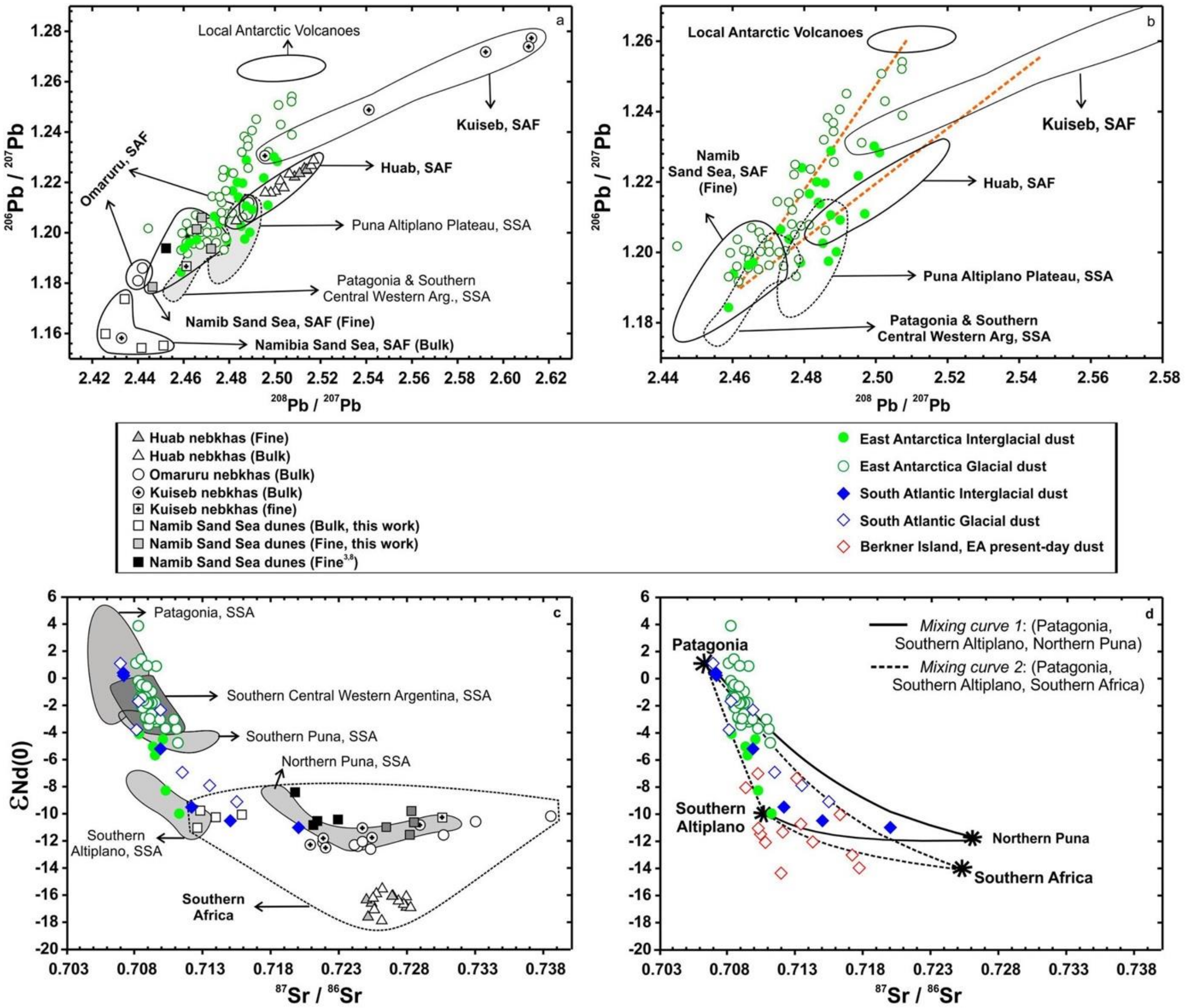

Fig. 14: Comparison of Southern Africa (SAF) and Southern South America (SSA) isotopic signatures with the glacial/interglacial fingerprint of dust deposited in East Antarctica and the South Atlantic sector of the Southern Ocean. a, b ^{208}Pb/^{207}Pb vs. ^{206}Pb/^{207}Pb. The orange dotted lines in Fig. 6b represent compositional trend lines necessary to explain EA dust compositions. c, d ^{87}Sr/^{86} Sr and εNd(0). The black lines in Fig. 6d represent the calculated tri-mixing hyperbola between different PSAs of SAF (dotted line) and SSA (fill line). End-member values used to calculate Sr–Pb mixing curves and references can be found in Gili et al. (2022).

In summary, this study confirms that SSA is the best candidate to explain the dust signature recorded in the SO and East Antarctic during cold and warm stages. For warm periods, this study proposes a scheme where the typical isotopic variability observed in interglacial and present-day dust depositions in the South Atlantic Ocean and coastal regions of East Antarctica can be related to the contributions of two main PSAs: Southern South America, through westerlies dominating the atmospheric circulation over Patagonia and Southern Central Western Argentina and, the jet stream over the Puna–Altiplano Plateau, in combination with Southern Africa (in response to the plausible transport by strong orographic winds (Gili et al., 2022).

4.2.4 Conclusions and Recommendations

Passive sampling set up could be improved

Exploratory investigations ahead of the CHASE project were possible thanks to team members of ULB during earlier expeditions to Princess Elisabeth station (Jean-Louis Tison and Vinciane Debaille), which were very useful to evaluate the snow amount to be collected and to test the Sigma 2 passive sampling system. Although special attention was paid on the narrow and staggered openings of the Sigma-2 to protect the inner part of the Sigma-2 from wind, solar radiation and precipitation, Sigma-2 passive samplers can be strongly impacted by snow accumulation inside the sampler, snow freezing, melting and sublimation. The snow can seal the sampler apertures during an undetermined time interval, with potential interruption(s) throughout the dust sampling period. The snow or ice deposited on the filter can melt, inducing salt accumulation on the filter and a partly loss of the particles. The savilex® vial placed in 2018 solved this problem. Eventually, the high-density PVC material used for the device was suitable and not damaged during the successive seasons.

Surface snow sampling seems to be a better solution for an efficient and faithful passive dust sampling. Moreover, the data compilation, as a whole, shows no significant differences between results obtained by the Sigma-2 filters relative to the snow samples. The best alternative could be using active dust sampling. That technique should be considered in the future despite additional severe logistical issues due to meteorological conditions in Antarctica. However, it induces a major difference compared to a passive sampling as it does not concern the study of dust deposition, but the study of the suspended aerosol particles in the air.

Preserving clean air around Antarctic research stations

The preliminary results obtained from the active samplers placed in the "atmosphere" container near the PEA station were useful, but also reveal significant contribution of atmospheric particle or component emissions from the station. The measured levels absolutely bear no air quality risk around the station, but are too high to perform high resolution analyses of the real background, pristine Antarctic atmospheric composition. However, because high-volume sampling needs strong pumps with high power demand, it is absolutely necessary to proceed and progress with the capacity and durability of zero-emission operation, including power generation, of Antarctic research stations. At PEA a lot of progress on this way has been made. Further innovations have to be investigated and tested, like zero-emission vehicles or local production of hydrogen for power-provision.

Ice core sites for paleoclimate reconstruction and future climate assessment

The methodology for identifying potential source areas (PSA) for the deposition of aeolian dust demonstrated by the CHASE project could also be applied to Aeolian dust deposits in Antarctic ice cores. This would be instrumental not only to reconstruct the paleoclimate in the southern hemisphere continental source regions, but also to improve estimates of the dust climate feedback through iron fertilization. Establishing a record of dust provenance would also help to better constrain shifts in the position and strength of the southern hemisphere westerly winds, which is key to our understanding of atmospheric and oceanic circulation changes during the last glacial cycle.

4.3 Source region and transport pathways analysis of atmospheric particles and VOCs

4.3.1 Source regions and atmospheric transport pathways and their seasonal variation

The FLEXTRA model has been successfully applied to calculate air mass trajectories and a k-means cluster analysis has been done. When the clustering is performed relying on the normalised latitude, longitude and altitude, four clusters of air mass origin were found. Figure 15 below shows the four air mass origin clusters for the austral summer season (December-January-February) and autumn season (March-April-May). Similar, in Figure16, the four air mass clusters are shown for austral winter (June-July-August) and spring (September-October-November).

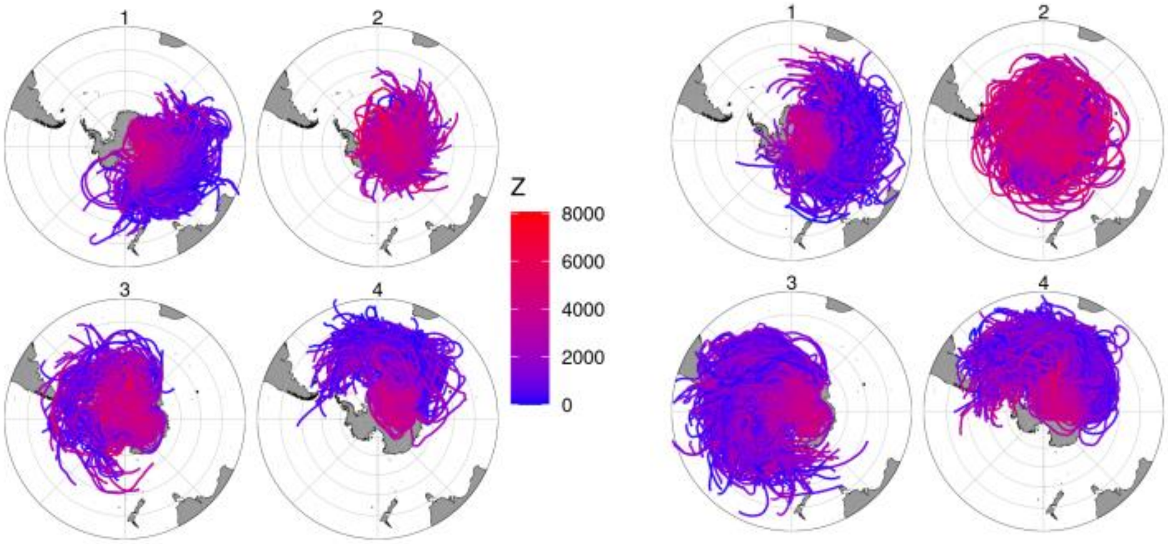

Figure 15: clusters of air mass origin, for austral summer (left) and austral autumn (right), for 11 years of backward trajectories (2010-2020) starting at PEA station; Z is height in m asl. All 10 days of the back trajectory calculation have been used for the clustering and all 10 days are included in the graphs

Some distinct features can be seen in the air mass origin clustering. Source regions from South America, Southern Africa and Australia were very limited. The Southern Ocean was a main source region, as was the Antarctic continent itself. For one of the clusters, the source region is mostly restricted to the region above the Antarctic continent. The average altitude along the trajectories in this cluster is higher compared to the average altitude of air coming from source regions over the Southern Ocean, indicating that this cluster corresponds to air subsiding from aloft.

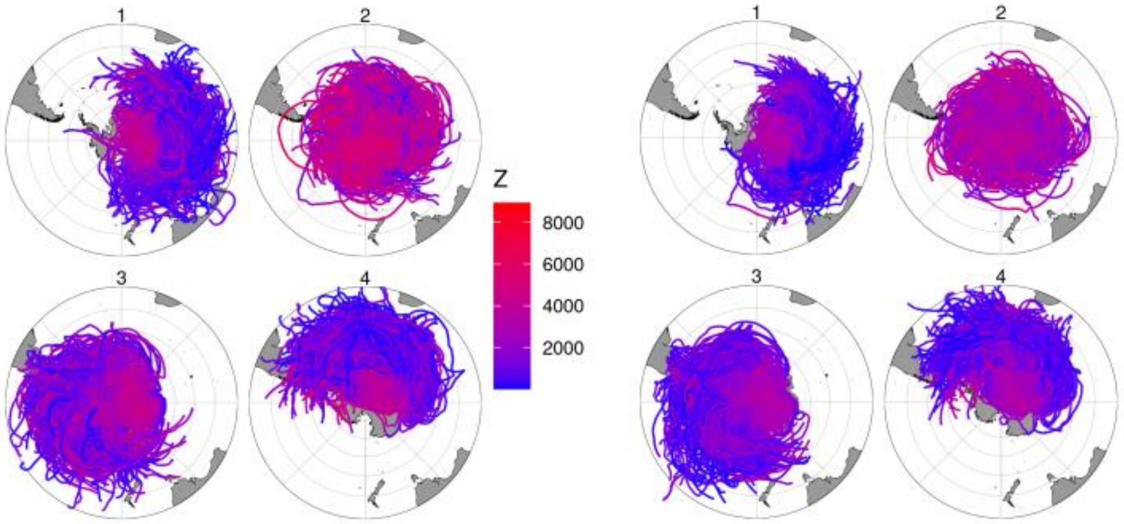

Figure 16: clusters of air mass origin, for austral winter (left) and austral spring (right), for 11 years of backward trajectories (2010-2020) starting at PEA station; Z is height in m asl. All 10 days of the back trajectory calculation have been used for the clustering and all 10 days are included in the graphs.

It is interesting to look at to what extent each cluster contributes to the total of backward trajectories per season. The percentages are given in Table 3. It can clearly be seen that in each season, cluster 2, representing mainly air masses of continental origin, with a large influence of upper tropospheric air, dominated. This indicates that the region of PEA was most of the time influenced by air masses transporting Antarctic particle background concentrations.

Table 3: percentages of backward trajectory occurrence, per air mass cluster and season; DJF = December-January-February; MAM = March-April-May; JJA = June-July-August; SON = September-October-November

Season	**Cluster 1**	**Cluster 2**	**Cluster 3**	**Cluster 4**
DJF	18 %	54 %	18 %	10 %
MAM	20 %	37 %	20 %	23 %
JJA	25 %	31 %	20 %	24 %
SONs	19 %	42 %	19 %	20 %

Figure 17 shows the probability density of the measured mean particle number concentration within the four air mass clusters and the four seasons. Some distinct differences among the seasons and among the clusters can be seen. For example, for the winter season, cluster 1 showed the highest concentrations and cluster 3 the lowest mean particle number concentration.

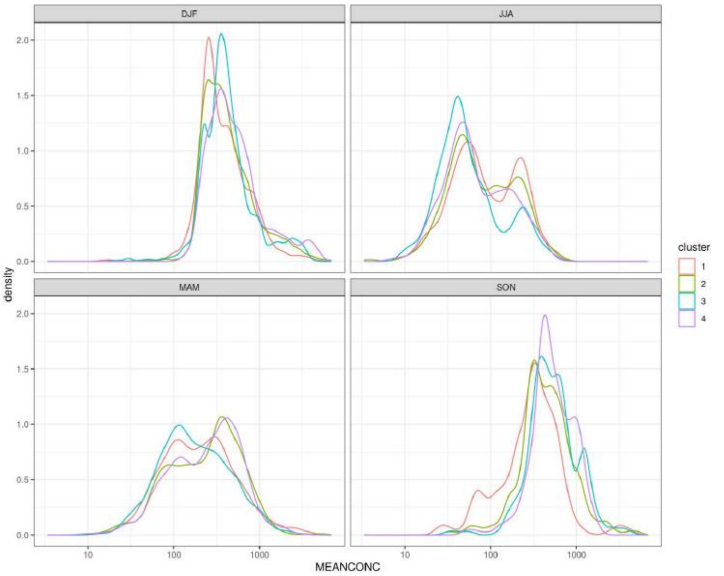

Figure 17: probability density distribution for measured particle number concentration, for the four seasons and the four air mass clusters, respectively

Figure 18 shows the probability density of potential vorticity (PV) within the four air mass clusters and the four seasons. PV is an indicator for air masses influenced by the stratospheric atmospheric circulation. Again, some distinct differences among the seasons and among the clusters can be seen. For cluster 2, the source region is mostly restricted to the region above the Antarctic continent and the average PV along the trajectories in this cluster is higher compared to the average PV within the other clusters, indicating that cluster 2 corresponds to air subsiding from aloft.

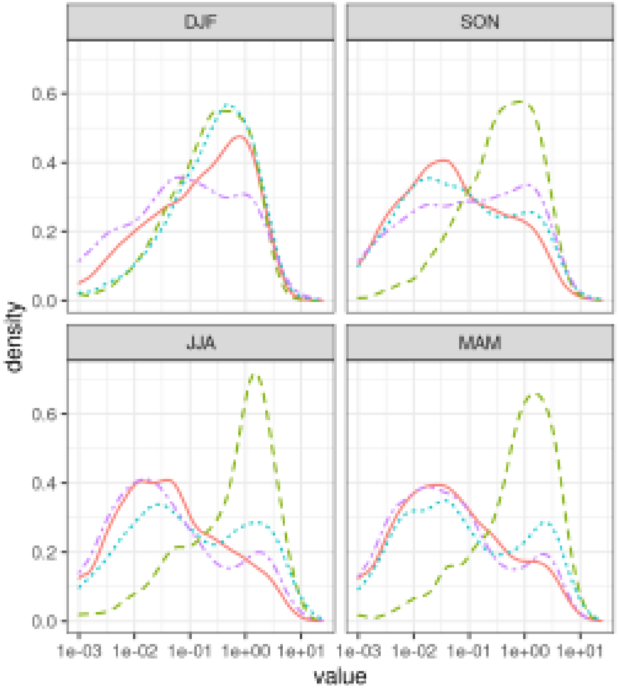

Figure 18: probability density distribution for potential vorticity along the calculated backward trajectories, for the four air mass clusters and the four seasons, respectively

Further, the FLEXPART dispersion model has been applied in order to identify potential source regions for the individual sample periods of the active sampling during seasons 2017/18 up to 2020/21. Figure 19 shows as an example the source regions for two measurement periods of the active sampling.

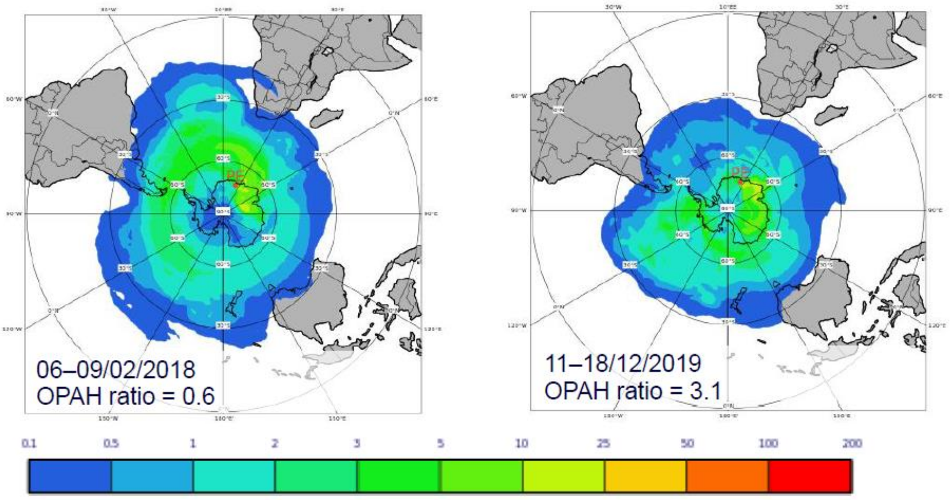

Figure 19: source regions for two periods of active filter sampling; the colour code gives the probability that the air mass has passed through that region

From Figure 15 it can be seen that for the sample with higher ratio of oxygenated PAHs, the air masses spent longer over the continent. For the sample with a low ratio of oxygenated PAHs, the air masses spent much more time over the Southern Ocean.

4.3.1 Implications of found particle chemistry on cloud condensation nuclei and ice nuclei

Figure 20 shows number concentrations of atmospheric particles, measured at PEA. From the total particle number it is clear that the overall amount of particles in the Antarctic atmosphere is very low (by a factor of around 10-100 lower than in Europe for example). The comparison of particle number between total number and number of particles of size ranges above around 100 nm demonstrates that most of the particles (around 80-90 %) are particles smaller than around 100 nm of diameter. This indicates a strong scarcity of available particles for cloud condensation nuclei (CCN) and ice nuclei (INP) because such particles are mostly of sizes of several hundreds of nm. These observations give already an indication of the available potential number of CCN and INP.

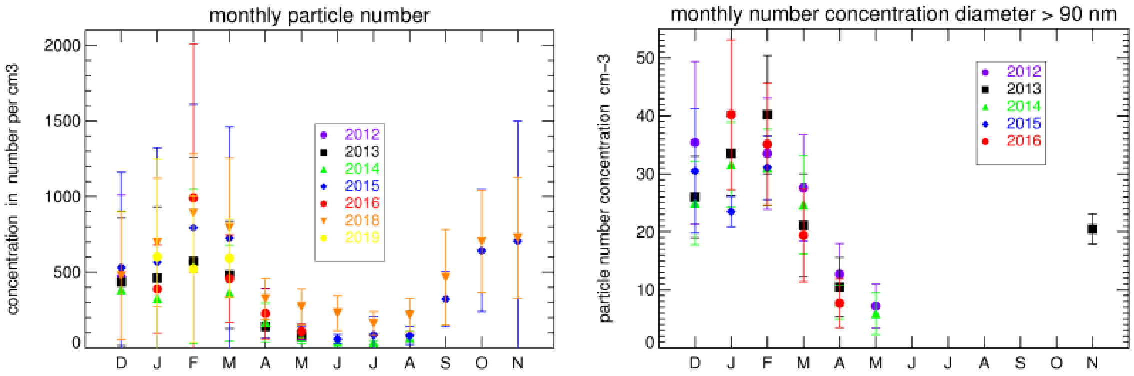

Figure 20: multi-annual monthly means of aerosol in-situ physical properties at PEA: total atmospheric particle number concentration (3-3000 nm; left) and particle number concentration of larger particles (> 90 nm size)

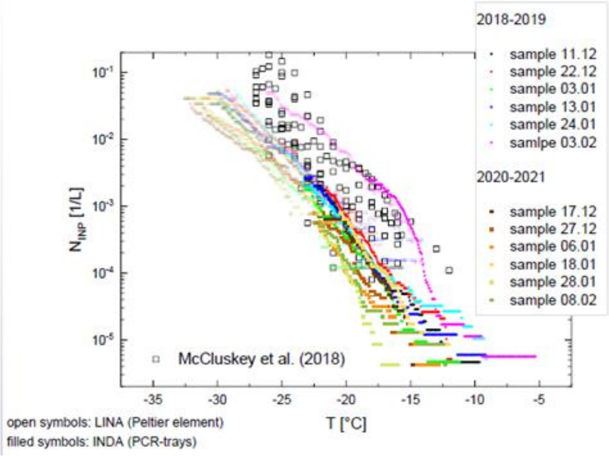

Figure 21: results of the laboratory analysis of INP filter samples taken during field seasons 2018/19 and 2020/21; analysed by TROPOS

Figure 21 shows results of the analysed INP filters taken at PEA during the field seasons 2018/19 and 2020/21. At PEA, low INP concentrations were obtained. Compared to the scarce literature data, the INP numbers for PEA are at the lower limit. This is an important finding, particularly as INP play an important role in ice formation in clouds and hence in precipitation formation. Studies indicate that the role of changes in INP for clouds dominates over the role of changes in CCN (Solomon et al., 2018, results of the BRAIN-be AEROCLOUD project, Souverijns, 2019). This information will be useful for the parameterisation of cloud and precipitation formation within (regional) climate models for Antarctica.

4.3.3 Recommendations

The results of the FLEXTRA and FLEXPART model calculations demonstrated that these are useful tools to find structures in the atmospheric transport pathways to Antarctica. Our results show that air masses of Antarctic continental origin dominate the potential source regions, followed by the Southern Ocean. Contributions from other continents (southern South America, southern Africa, Australia) were found to be marginal. However, as by the chemical analyses signatures were found which can clearly be attributed to certain source regions (e.g., via the REE elemental analysis) or are of anthropogenic origin (some VOCs, PAHs), there are certainly pathways of these compounds into

East Antarctica. But respective simulations of current atmospheric transport models have large uncertainties when simulating several weeks of atmospheric transport what seems to be necessary for transport into Antarctica. Therefore, more elucidated modelling and research is needed how the East Antarctic atmosphere is connected to lower latitudes.

Further, the found low concentrations of ice nucleating particles show the need to obtain more such measurements, in particular because INP play an important role in ice formation in clouds and hence in precipitation formation (what is the only source term for the Antarctic surface mass balance).

5. DISSEMINATION AND VALORISATION

5.1 Network management

5.1.1 Project coordination

Project coordination has been led by partner RMI. Throughout the course of the project, several meetings were organised bringing together all partners of the CHASE project. Meetings were in-situ before Covid19-related restrictions and online afterwards. At least one meeting per year was especially dedicated to discuss the scientific progress and collaboration between the partners. Ahead of each field expedition to PEA, a meeting took place in order to prepare the campaign and another meeting was hold afterwards for debriefing the field expedition. Apart from this, ad-hoc meetings took place to discuss current scientific matters or campaign planning, not necessarily involving all partners. Several of the publications listed in section 6 (Publications) are based on the collaboration between the different partners of the CHASE project and have partners from the different institutes as co-author.

5.1.2 Interaction with Follow-up Committee

The progress of the CHASE project has been discussed with the members of the follow-up committee. Here, we describe the interaction with them during the project.

- Nadine Mattielli kept contact with Profs. Karine Deboudt and Pascal Flament **(**Laboratory of Physics and Chemistry of the Atmosphere (LPCA), Université du Littoral – Côte d'Opale, Dunkerque, France). They provided their expertise in aerosol characterisation by applying single-particle analysis (SEM-EDX) on the suspended atmospheric particles collected directly on filters (Sigma-2 and active pump samplers) and dust deposits (snow samples).
- Nadine Mattielli was in contact with Dr. Volker Dietze (German Meteorological Service, Research Centre Human Biometeorology, Air Quality Department, Freiburg, Germany) who provided the passive sampling equipment for inorganics. They discussed the installation and improvement of the samplers.
- Christophe Walgraeve met with Dr. Willy Maenhaut (former Dept of Analytical Chemistry, Ghent University, Belgium) who is a reknown international expert on aerosol chemistry and measurement techniques and has performed aerosol chemistry measurements at the South Pole. They discussed the practical details of the setup of the active filter sampling.
- Alexander Mangold has been in discussions with Rolf Weller (Alfred Wegener Institute, Bremerhaven, Germany) on data of the atmospheric aerosol in-situ physical properties.
- Alexander Mangold has been and is in contact with Prof. Nicole Van Lipzig (KU Leuven, Belgium). Within the Brain-Be Aerocloud project they collaborated on investigating the relationship between clouds, precipitation and aerosols in Antarctica. This collaboration is continued within the CLIMB BrainBe project. Within her group, the COSMO-CLM2 regional climate model has been adapted to simulate also the influence of different types of particles on the formation of clouds and precipitation.
- Preben van Overmeiren collaborated with Prof. Laszlo Vincze Ghent University, Belgium), his former MSc supervisor. Prof Vincze is doing research on (interstellar) dust characterisation

using Synchrotron X-Ray analysis. Together with him first analyses have been made on collected Antarctic samples. However, the results were not promising enough in order to further explore this possibility.

- CHASE partners are in contact with Prof Annick Wilmotte_(University of Liège, Belgium). Her group and collaborators are studying the microbial diversity on deglaciated rocks, nunataks, or ridges in Antarctica. They are interested in how such taxa are distributed in Antarctica, e.g. via air transport. Our filter material might therefore have been useful for microorganisms analyis. First test filter material has been exchanged with her. In addition, she is member of the Belgian delegation to the Committee for Environmental Protection to the Antarctic Treaty. She has prepared a document to protect some of the biological soil crusts in the Sor Rondane mountains and to keep a reference area non-violated for future studies. We discussed with her potential input (arguments, areas) with respect of our needs to sample non-disturbed atmospheric particles or re-distributed soil dust and gave her information on the meteorological conditions at these areas.

5.1.3 Maintenance of observatory at Princess Elisabeth station

In order to gain reliable results, the instrumentation deployed for CHASE purposes had to be maintained properly. Ahead of each austral summer expedition to PEA, the following preparations were undertaken:

- In-situ and virtual (due to Covid19 limitations) meetings and email-exchanges with the station operator and between CHASE partners, in order to discuss the practical topics for the field campaigns;
- Administrative organisation of the campaign in cooperation with the polar secretariat and the station operator, including shipment formalities;
- Equipment for the filter sampling and snow sampling for chemical analyses has been cleaned and prepared for assuring non-contaminated samples;
- Preparation, maintenance and if needed repair of instrumentation for the auxiliary measurements (aerosol in-situ, radio soundings)

Each austral summer season, staff from the CHASE partners was present at PEAS and carried out the collection of samples and instrument operations. The following gives a brief description of the individual field expeditions:

- Season 2017/2018: 3 Pax present November-December 2017 (Alexander Mangold (on AEROCLOUD project), Nadine Mattielli, Christophe Walgraeve); installation of active sampling and of first passive sampling sites
- Season 2018/2019: 2 Pax present November-December 2018 (Stefania Gili and Preben Van Overmeiren); re-installation of active sampling and completing transect of passive sampling sites, collection of samples;
- Season 2019/2020: 2 Pax present November-December 2019 (Stefania Gili and Preben Van Overmeiren); re-installation of active sampling and maintaining passive sampling sites; collection of samples;

- Season 2020/2021: 2 Pax present November-December 2020 and 1 Pax until mid-January 2021 (Preben Van Overmeiren and Alexis Merlaud); re-installation of active sampling and maintaining passive sampling sites; collection of samples;
- Season 2021/2022: 2 Pax present November-December 2021 (Preben Van Overmeiren and Andy Delcloo); de-installation of active sampling and of most passive sampling sites; collection of samples.

5.2 Data base and data dissemination

Most of the gathered data within CHASE concern chemical analyses from taken samples (filters, snow). It is in the nature of the chemical laboratory analyses that the original samples themselves are used up during these processes. The analytical results will be made public either after publication in scientific journals or along with the publications. The VOC dataset can possible be submitted to the EBAS database; https://ebas.nilu.no/). Data of the radio soundings has been sent to the Global Telecommunications System (GTS), data of the automatic weather station is available via partner RMI or from IMAU Utrecht, data of atmospheric aerosol physical properties will be delivered to the EBAS data base. Data for the atmospheric back trajectory and dispersion modelling is available via partner RMI (huge data amount, stored on RMI servers).

5.3 Dissemination of results to scientific community, stakeholders and the general public

Results of CHASE have been widely disseminated and have been made public. In particular, see sections 6.1 for peer-reviewed scientific publications and 6.2 for presentations at scientific conferences, and sections 6.3 Outreach to scientific community and stakeholders, and section 6.4 Outreach to the general public.

Further, the connection between scientific research in Antarctica and policy is largely managed by the Scientific Committee on Antarctic Research (SCAR). Belgium is full member of SCAR, represented by the Belgian National Committee on Antarctic Research (BNCAR). Prof. Philippe Claeys and Dr. Alexander Mangold are members of BNCAR and have been attending the meetings to ensure that all BNCAR scientists involved are aware of the on-going research. Also, CHASE partners are members of the SCAR Action Group ImPACT (Input pathways of persistent organic pollutants to Antarctica). This Action Group aims to facilitate coordinated investigation and monitoring of chemical input to the Antarctic region and aims to serve both the Global Monitoring Plan of the Stockholm Convention on Persistent Organic Pollutants (POPs), as well as the Protocol on Environmental Protection to the Antarctic Treaty (the Madrid Protocol) which explicitly prohibits the importation of POPs (chemicals of known risks) to Antarctica.

Initially, a scientific workshop was foreseen towards the end of the CHASE project. Unfortunately, due to the tight agendas of partners no suitable date could be found. However, CHASE partners participated in a polar symposium on 22 September 2022, organised by Belspo, the Egmont institute, BNCAR and APECS (Association of Polar Early Career Scientists). Results from CHASE have been presented there.

In addition to the contacts with the members of the Follow-up Committee (see section 5.1.2), CHASE partners have discussed the project and its outcomes with the scientific community as listed below:

- A collaboration of partner ULB with Paola Formenti, Senior Scientist of the National Center for Scientific Research (CNRS) at LISA (France), opened new perspectives on a better understanding of the dust genesis from the main dust precursors (soils or loess from Southern South America or South Africa).
- Nadine Mattielli collaborates with Prof. James King from Université de Montréal (Canada). He shared samples from his previous campaigns in Namibia.
- Barbara Delmonte, from the Department of Environmental and Earth Sciences (University of Milano-Bicocca, Milan, Italy) provided to partner ULB additional samples from southern Africa for new Sr, Nd and Pb isotopic analyses performed at the G-Time Laboratory.
- Nadine Mattielli collaborates with Prof. Steeve Bonneville, and G. Laruelle (from B-GeoSys, ULB) for the development of the statistical tools and the code required for the REE model set up. To have access to the REE data of prestigious Antarctic ice cores, such as EDML and EDC, N. Mattielli is in contact with Paolo Gabrielli (Ohio state University, USA) and Alois Bory (from Université de Lille, France), who positively reply for a fruitful collaboration.
- Thanks to Stefania Gili of partner ULB, the collaboration with Prof. Diego Gaiero_(National University of Cordoba, Argentina) was effective through common publications.
- In September 2020, Stefania Gili was hired as senior scientist at Princeton University, USA. She is in charge of the analytical laboratory and ice core projects. The close collaboration between Nadine Mattielli and Stefania Gili will be pursued and extended.
- Alexander Mangold collaborates with Dr. Heike Wex (Leibniz Institute for tropospheric research, TROPOS, Leipzig, Germany) who is doing research on cloud formation, cloud processes and the aerosol particles involved in it. Her group is interested in the chemistry of the particles sampled within CHASE.
- Nadine Mattielli and Alexander Mangold are in contact with Prof François Fripiat of ULB (Glaciology department). He is interested in the atmospheric chemistry of reactive nitrogen in Antarctica. The snow and especially the snow/ice core samples of CHASE are of interest to him, not only to potentially use them for his analyses but also to compare results of reactive nitrogen with the other chemistry results.

6. PUBLICATIONS

6.1 peer-reviewed scientific publications

- Gili, S., Vanderstraeten, A., Chaput, A., King, J., Gaiero D. M., Delmonte, B., Vallelonga, P., Formenti, P., Di Biagio, C., Cazanau, M., Pangui, E., Doussin, J-F. & Mattielli, N. (2022). South African dust contribution to the high southern latitudes and East Antarctica during interglacial stages. *Nature, Communications Earth & Environment*, https://doi.org/10.1038/s43247-022-00464-z
- Held, A. and Mangold, A. (2021), Measurement of Fundamental Aerosol Physical Properties, in: Foken, T. (ed.), Handbook of Atmospheric Measurements, Springer Handbooks. Springer, Cham, 533-563. https://doi.org/10.1007/978-3-030-52171-4_18
- Herenz, P. H. Wex, A. Mangold, Q. Laffineur, Gorodetskaya, I. V., Z. Fleming, M. Panagi and F. Stratmann (2019), CCN measurements at the Princess Elisabeth Antarctica Research station during three austral summers, Atmos. Chem. Phys. 19, 275-294, doi.org/10.5194/acp-19-275-2019, 2019.
- Maters, E., Mulholland, D., Flament, P., de Jong, J., Mattielli , N., Deboudt, K., Dhont, G., Bychkov, E. (2022), Laboratory study of iron isotope fractionation during dissolution of mineral dust and industrial ash in simulated cloud water. *Chemosphere*, 299, 134-472
- McCutcheon, J; Lutz, S.; Williamson, C.; Cook, J. M; Tedstone, A. J; Vanderstraeten, A.; Wilson, S. A; Stockdale, A.; Bonneville, S., Anesio, A.M., Yallop, M.L., McQuaid, J.B., Tranter, M. and L.G. Benning (2021), Mineral phosphorus drives glacier algal blooms on the Greenland Ice Sheet. *Nature Communications* 12 :1-11, 2021.
- Vanderstraeten, A., Bonneville, S., Gili, S., de Jong, J., Debouge, W., Claeys, Ph., Mattielli , N. (2020), First multi-isotopic (Pb-Nd-Sr-Zn-Cu-Fe) characterization of dust reference materials (ATD and BCR-723): a multi-column chromatographic method optimized to trace mineral and anthropogenic dust sources. *Geostandards and Geoanalytical Research*, vol. 44, issue 2, 307-329.
- Vanderstraeten, A. (2020), Advances in geochemical tracing for atmospheric dust in Antarctica, ULB PhD Thesis, September 2020; https://difusion.ulb.ac.be/vufind/Record/ULB-DIPOT:oai:dipot.ulb.ac.be:2013/312773/Holdings
- Vanderstraeten, A., Mattielli , N., Laruelle, G. G., Gili, S., de Jong, J., Bory A., Gabrielli, P., Boxho, S., Tison, J-L., Bonneville, S. (submitted). Identifying the provenance and quantifying the contribution of dust sources in Epica Dronning Maud Land over the last deglaciation: a high-resolution, continuous record from a new rare earth element mixing model. *Earth-Science Reviews* Journal, submitted.

6.2 Contributions to scientific conferences

6.2.1 Oral presentations

- De Causmaecker, K., A. Mangold, C. Walgraeve, P. Van Overmeiren, N. Mattielli, S. Gili and A. W. Delcloo, Validating aerosol plume data using satellite data and dispersion modeling, EUMETSAT Meteorological Satellite Conference , Bucarest, Romania, 20-24 September, 2021.
- De Causmaecker, K., A. Mangold, C. Walgraeve, P. Van Overmeiren, N. Mattielli, S. Gili, and A. W. Delcloo, Identifying source regions at the Princes Elisabeth station in Antarctica, using dispersion modelling tools: a case study, European Geosciences Union General Assembly 2020 (online video conference), Abstract EGU2020-9127, 4-8 May 2020, Vienna, Austria, 2020.
- Gili, S., Vanderstraeten, A., M. Cazaunau, A. Chaput, J.-F. Doussin, C. Di Biagio, P. Formenti, J. S. King, A. Mangold, N. Mattielli, E. Pangui, P. Van Overmeiren and C. Walgraeve, The role of Southern Africa as a dust precursor to East Antarctica, European Geosciences Union General Assembly 2020 (online video conference), Abstract EGU2020-18441, 4-8 May 2020, Vienna, Austria, 2020.
- Mangold, A., Q. Laffineur, A. Delcloo, V. De Bock, C. Hermans, F. Hendrick, P. Herenz, H. Wex, A. Gossart, N. Souverijns, N. van Lipzig, I. Gorodetskaya, and H. De Backer, Atmospheric aerosol characterisation and relations to clouds and precipitation in Dronning Maud Land, East Antarctica, European Aerosol Conference 2019 Abstract O10_F1_A05, 25–30 August, Gothenburg, Sweden, 2019.
- Mangold, A., Q. Laffineur, A. Delcloo, C. Hermans, F. Hendrick, A. Gossart, N. Souverijns, P. Herenz, H. Wex, N. Van Lipzig and H. De Backer, Aerosol variability linked to clouds and precipitation in the Sor Rondane area, POLAR2018, SCAR/IASC Open Science Conference, Davos, Switzerland, 19-23 June 2018.
- Mangold, A., Q. Laffineur, H. De Backer, A. Delcloo, P. Herenz, H. Wex, A. Gossart, N. Souverijns, I. Gorodetskaya, N. Van Lipzig, CCN and aerosol properties at Princess Elisabeth station, East Antarctica, combined with cloud and precipitation observations and air mass origin, European Geosciences Union General Assembly 2017, 23 - 28 April 2017, Vienna, Austria.
- Vanderstraeten, N. Mattielli, G. G Laruelle, A. Bory, S. Gili, P. Gabrielli, S. Boxho and S. Bonneville. High-resolution statistical quantification of aeolian dust provenance in East Antarctica over the Last Glacial-Interglacial Transition. Abstract Goldschmidt Conference, 4-9 July 2021, Lyon, France.
- Vanderstraeten, A., S. Gili, A. Mangold, S. Bonneville, V. Dietze, C. Walgraeve, P. Van Overmeiren, C. Berclaz, S. Goderis, N. Mattielli, Chasing dust in Dronning Maud Land, East Antarctica: A trace element perspective, 20th Congress of the International Union for Quaternary Research (INQUA), Dublin, Ireland, 25-31 July, 2019.
- Van Overmeiren P., Gili S., Mattielli N., Mangold A., Demeestere K., Van Langenhove H., Walgraeve, C., Organic compounds in the Antarctic atmosphere : first results of the 2017 and 2018 field sampling campaign, ‘Antarctic climate symposium’, Belgian Science Policy BRAIN-be Aerocloud event, Brussels, 10 May 2019.

- Van Overmeiren, P., S. Gili, A. Vanderstraeten, N. Mattielli, A. Delcloo, K. De Causmaecker, A. Mangold, K. Demeestere, H. Van Langehove, C. Walgraeve, Obtaining insight in atmospheric trace organic compound concentrations and trends in Dronning Maud Land, East Antarctica by means of long term passive and active air sampling, SCAR Open Science Conference (only online), Hobart, Australia, 3-7 August 2020.
- Van Overmeiren, P., A. Delcloo, K. De Causmaecker, A. Mangold, K. Demeestere, H. Van Langenhove and C. Walgraeve, Sequential sampling of Volatile Organic Compounds (VOCs) and atmospheric oxidation products in the Sor Rondane Mountains, East-Antarctica, EGU General Assembly 2022, Vienna, Austria, 23 May 2022.

6.2.2 Poster presentations

- De Bock, V., H. De Backer, A. Mangold, Q. Laffineur and A. Delcloo, UV measurements at Uccle, Belgium (1990-2018) and Utsteinen, Antarctica (2011-2018), European Conference on Solar UV Monitoring, Vienna, Austria, 12.09.-14.09.2018.
- Mangold, A., K. De Causmaecker, A. Delcloo, Q. Laffineur, P. Van Overmeiren, C. Walgraeve, K. Demeestere H. Van Langehove, S. Gili, A. Vanderstraeten and N. Mattielli, Climatology of air mass origin for Princess Elisabeth Antarctica station: clustering and analysis for atmospheric particle properties and semi-Volatile Organic Compounds, European Aerosol Conference 2021 (online) Abstract 200-AAS-P2, 30 August – 3 September 2021, Manchester, UK, 2021.
- Mangold, A., H. De Backer, V. De Bock, K. De Causmaecker, A. Delcloo, Q. Laffineur, F. Hendrick, C. Hermans, P. Herenz, H. Wex, P. Van Overmeiren, C. Walgraeve, S. Gili, and N. Mattielli, Atmospheric aerosol in Dronning Maud Land, East Antarctica: physical and chemical properties and source region analysis, European Aerosol Conference 2020 (online) Abstract P3-036, 30 August – 4 September, Aachen, Germany, 2020.
- Mangold, A., Q. Laffineur, A. Delcloo, V. De Bock, C. Hermans, P. Herenz, H. Wex and H. De Backer, Atmospheric Aerosol Characterisation at Princess Elisabeth Station, Antarctic Climate Symposium, Belgian Science Policy BRAIN-be Aerocloud event, Brussels, 10 May 2019.
- Mangold, A., Q. Laffineur, R. Van Malderen, C. Hermans, K. Nys, M. Verbruggen and H. De Backer, Total ozone, UV and radio sounding measurements in the Sør Rondane Mountains, POLAR2018, SCAR/IASC Open Science Conference, Davos, Switzerland, 19-23 June 2018
- Mangold, A., Laffineur, Q., De Backer, H., De Bock, V., Delcloo, A., Hermans, C., Gielen, C., Herenz, P., Wex, H. (2017). Atmospheric aerosol and CCN properties in Dronning Maud Land, East Antarctica, European Aerosol Conference 2017, Zürich, Switzerland.

6.3 Outreach to the scientific community

- Gili, S. *Chasing dust in Dronning Maud Land, East Antarctica: A geochemical perspective*. Seminar, Ghent University, 10 May 2019
- Gili, S., chaired the session during the International Union for Quaternary Research (INQUA), Dublin, Ireland, 25-31 July, 2019
- Mangold, A., Measurements of atmospheric particles in Antarctica: what even a low number of particles can tell us about the Antarctic atmosphere, Nanoparticles and air quality (talk in French), Seminar of TSI, JJBos, 21 May 2019, Ottignies-Louvain-La-Neuve, Belgium
- N. Mattielli chaired the session *Mineral Dust In The Earth System* at the international Goldschmidt conference, hold online, Lyon, July 2021.
- Contribution to the Nanok project (CDR – FNRS): *Phosphorus and Source of Greenland Dust* (PI: Steeve Bonneville; project members: Sibylle Boxho, Nadine Mattielli, & Aloys Bory (Université de Lille, France); https://www.nanokexpedition.be
- Vanderstraeten, A., PhD Thesis – Advances in geochemical Tracing of Atmospheric Dust in Antarctica, ULB, public defense in September 2020, 255p.
- Van Overmeiren, P., Ghent University Faculty Research Days, Unraveling atmospheric chemistry in Antarctica, Faculty of Bioscience engineering, Ghent University, 5 May 2021.

6.4 Outreach to the general public

Lectures

- Presentation on the usage of sustainable energy in science by Preben Van Overmeiren during an EU Green Week partner event hosted by the International Polar Foundation, Schneider Electric and Venturi, 30 May, 2022
- Every year during the Belare expedition, Preben Van Overmeiren gave a short lecture (+-30min) about the science covered by the CHASE project to the staff of IPF deployed at the station.
- Every season during the Belare expedition, Preben Van Overmeiren gave video-chats from the PEA station to secondary schools in Belgium, e.g., short lecture on life and science on Antarctica from the PE Station to the 5th and 6th year of the municipal primary school 'De Droomwolk' (Beveren-Waas) 14-01-2021.
- Mangold, A., Antarctica and Climate – what it is about, Asgard IX, European Space Education Resource Office, weather balloon launch competition for schools, talk and discussion with students from international secondary schools, 25 April 2019 Uccle, Belgium.

Media presence (print, website, interviews)

- A reporter from the Belgian newspaper 'Het Laatste Nieuws' interviewed Preben Van Overmeiren before departing for the second field campaign 2017/18. The article was published on 16/11/2018

- http://www.antarcticstation.org/news_press/news_detail/preben_van_overmeiren_explains_aerosols_and_cloud_formation__in_antarctica_ (9 February 2021); short version also on UGent faculty website: https://www.ugent.be/bw/en/news-events/news/expedition-antarctica-atmospheric-particles-preben-van-overmeiren.htm
- http://www.antarcticstation.org/multimedia/picture_gallery/traverse_for_the_chase_and_climb_projects_ (9 February 2021);
- http://www.polarfoundation.org/news_press/news/stefania_gili_on_studying_atmospheric_particles_in_antarctica_ (1 September 2020);
- http://www.polarfoundation.org/news_press/news/alexander_mangold_contributions_of_research_to_polar_science__yopp_ipcc (22 July 2020)
- At the beginning of each Belare field expedition, RMI published on its main website a 'NEWS' (Actualité, 'In de kijker'); similar, Ghent University and ULB issued press releases or published respective news features on their websites.

Videos/Movies

- Prof. Nadine Mattielli presented on 9 February and 25 March 2020 the CHASE project in 'PITCH', a dedicated programme of the Brussels TV channel Bxl1 (https://bx1.be/emission/pitch-nadine-mattielli-ulb/)
- TV documentary of the field season 2019/20 at PEA, including interviews with Preben Van Overmeiren and Stefania Gili of CHASE and passages on their field work: https://www.siouxproductions.be/les-gardiens-de-lantarctique distributed on RTBF (public broadcaster, French-speaking; December 2020 and three months available via streaming: https://www.rtbf.be/tipik/article/detail_les-gardiens-de-l-antarctique-votre-nouvelle-serie-documentaire-debute-ce-lundi-sur-tipik?id=10634510) and likewise on VRT (public broadcaster, Flanders, dutch-speaking; March 2021)

Blogs:

- RMI maintains a continuous blog on RMI's activities at Princess Elisabeth station: https://belatmos.blogspot.com
- During field season Belare 2017/2018, Nadine Mattielli and Christophe Walgraeve wrote about their research activities and during Belare 2018/2019, Stefania Gili and Nadine Mattielli: www.bncar.be
- Blog by Preben Van Overmeiren on the research activities during Belare 2020/21 (https://ozone.meteo.be/projects/chase/belare2020-2021-campaign)
- Blog by Preben Van Overmeiren and Andy Delcloo on the research activities during Belare 2021/22 (https://ozone.meteo.be/projects/climb/belare-2021-2022-campaign-v2)

Other

- Scientific photography exhibition of photo's taken by Preben Van Overmeiren during the CHASE expeditions in the Royal Museum of Natural Science, Nocturnes on Ice organised by APECS Belgium, 19-22 May 2022

- Mangold, A., Notre l’environnement, le climat et l’Antarctique, Workshop for children of 12-13 years, presentation and experiments, TADA, (http://toekomstatelierdelavenir.com), 24 October 2020, Anderlecht, Belgium.
- Mangold, A., L’Antarctique, le pôle Sud et la Belgique, explaining to school children (8-9 years and 10-11 years) of the Ecole Communale de Linkebeek (BE) what Antarctica is, why research is done there and what are the conditions for work and life, 26 June 2018, Linkebeek, Belgium.

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