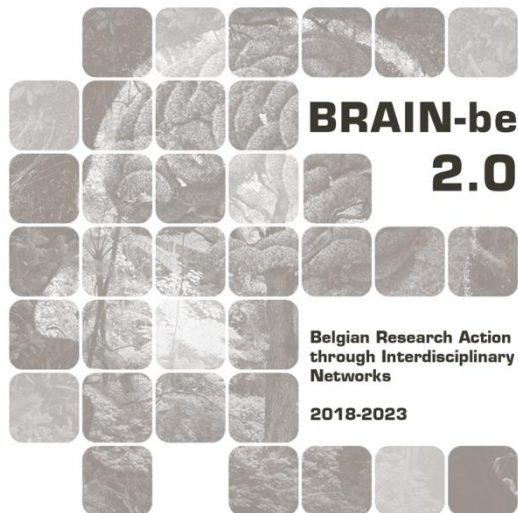


## **CLIMB**

### **How do aerosol-Cloud Interactions influence the surface Mass Balance in East Antarctica?**

Alexander Mangold (RMI) – Andy Delcloo (RMI) – Nicole van Lipzig (KUL) – Florian Sauerland (KUM) – Michel Van Roozendael (BIRA-IASB) – Alexis Merlaud (BIRA-IASB) – Martina M. Friedrich (BIRA-IASB) – Christophe Walgraeve (UGent) – Preben Van Overmeiren (UGent)

Pillar 1: Challenges and knowledge of the living and non-living world



NETWORK PROJECT

**CLIMB**

**How do aerosol-Cloud Interactions influence the surface Mass Balance in East Antarctica?**

Contract - B2/191/P1/CLIMB

**FINAL REPORT**

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## **ABSTRACT**

### **Context**

The water cycle, cloud microphysics and cloud-aerosol-interactions are recognized as key elements of the Antarctic climate system by several international consortia, such as the International Panel on Climate Change (IPCC). Clouds and aerosols play a significant role in the radiative energy budget and aerosols impact cloud microphysics because they are cloud condensation and ice nuclei. In addition, clouds are an important part of the hydrological cycle serving as the agent linking water vapour transport into Antarctica with precipitation. Because precipitation is the only source term in the surface mass balance (SMB) of the Antarctic ice sheet, it is one of the key factors affecting sea level. However, current knowledge on the interaction between clouds, precipitation and aerosol in the Antarctic is still limited, both from direct observations and from regional climate models.

CLIMB relied on an interdisciplinary research team, bringing together scientists with complementary expertise in long-term experimental work related to atmospheric chemistry and physics, expertise in Antarctic research campaigns, regional climate modelling, radiative transfer processes, and expertise in state-of-the-art chemical analyses.

CLIMB performed measurements of meteorological, aerosol, cloud and precipitation characteristics, both in the vicinity of Princess Elisabeth Antarctica station (PEA), and at an altitude which was high enough and exposed to easterly winds to be often within cloud level. In addition to extended ice nuclei particle (INP) filter sampling at PEA, there were (i) a vertically resolved profile of temperature, relative humidity and pressure for three heights: at PEA (1390 m asl), on the Utsteinen nunatak summit (around 1600 m asl) and in the Vikinghogda mountains (2350 m asl); (ii) measurements of precipitation type and intensity by a disdrometer; (iii) measurements of aerosol particle size in the Vikinghogda mountains; and (iv) an automated sampling system for Volatile Organic Compounds (VOC), also in the remote Vikinghogda mountains. The results were used to improving the aerosol-cloud-precipitation parameterisation in a regional climate model for East Antarctica.

### **Objectives**

The objectives of CLIMB were:

- to deliver a unique data set of in-cloud meteorological, aerosol and cloud characteristics, combined with simultaneous boundary layer and ground-based remote sensing measurements;
- to establish a data set with a detailed mapping of air mass origins and transport pathways into East Antarctica;
- to generate an improved COSMO-CLM<sup>2</sup> regional climate model for Antarctica;
- to improve the understanding of the climatological effect of cloud condensation and ice nuclei particles on clouds, precipitation, radiation and the surface mass balance in East Antarctica;
- to strengthen the role of Belgium in the Antarctic research community;
- to valorise the results by scientific publications and workshops, open data access, lectures to the general public and press contributions.

## Conclusions

- The aerosol optical depth was determined by both CIMEL sunphotometer and MAX-DOAS measurements. For three austral summer seasons, overlapping data exists. Typical AOD values in the UV and visible range of 0.02 to 0.04 were found, close to those previously reported in the literature for pristine Antarctica.
- Ground-based MAX-DOAS measurements were exploited to infer the abundance of stratospheric ozone, the NO<sub>2</sub> vertical column, and the OCIO slant column – key chemical species involved in the processes leading to the ozone hole. Measurements for 2022 showed the OCIO build-up during July-August, the polar stratospheric clouds induced denoxification/denitrification, and the corresponding ozone depletion, reaching a minimum total ozone value of 126 DU by end of October.
- A high variation of the temperature difference between the Utsteinen nunatak and ridge (around 200 m altitude difference) was found, higher than could be explained by the mere difference in height. This feature of local meteorology needs further investigation.
- The analysis of the cloud phase showed that liquid-containing clouds were much more frequent during austral summer, when in about a third of all clouds, liquid water has been detected, compared to other seasons. These events were found to correlate to more northerly winds in the upper atmosphere.
- The purpose-built automated sampler for VOCs proved capable of taking multiple samples from December 2019 to October 2020. This led to the first ever year-round dataset of 66 (oxygenated)VOCs reported for East-Antarctica and marks the first time that these compounds were successfully sampled in an automated sequential manner in Antarctica for later, off-line analysis. The laboratory analyses led to a dataset, spanning across different groups of organic compounds (like aromatic, halocarbons), from very reactive to very persistent, and molecular weights from 46 to 253 g/mol. Concentration ranges from 0.5 ng/m<sup>3</sup> (methylbenzoate) to 20 µg/m<sup>3</sup> (benzoic acid) were found. Almost half of the detected species in the samples were oxygenated compounds.
- During two austral summer seasons, samples were collected for the analysis on the concentration of ice nucleating particles (INP). Compared to studies in other regions of Antarctica, the found INP numbers for PEA are at the lower limit. This is an important finding, in particular for modelling studies on the aerosol influence on cloud formation and precipitation. The measurements of the optical particle sizer indicated that the concentration of particles larger than 500 nm (sizes relevant for INP) was very low. Overall, the Antarctic continent itself seemed to be no important contributor to INP.
- The regional climate model COSMO-CLM2 was adapted and improved to investigate the effect of INP on clouds and the climate of East Antarctica. The model was found to represent accurately cloud height and occurrence. A strong anti-correlation between INP concentration and liquid water content was found, including INP concentrations representative for the region around PEA. Overall, CLIMB results indicate that the exact concentration of INPs, as long as it is within a realistic range for the region, is not strictly required for a good model

performance. However, a low INP concentration caused an increase in the cloud radiative effect both for short- and longwave radiation. These opposing effects mostly cancelled out in the austral summer, while during the winter, the lack of incoming shortwave radiation meant a stronger warming effect through the increase in reflected longwave radiation.

- A climatology of backward air mass trajectories has been established 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. The results show that air masses of Antarctic continental origin dominated the potential source regions, followed by the Southern Ocean. Contributions from other continents were found to be marginal. Over all seasons, the Antarctic continent with air masses between 1400 m and 8000 m was the dominant source region, with mainly the altitude level between 1400 and 4000 m. The Antarctic continental source was most prominent during austral summer with more than 80% share of origin. Air masses with maritime origin had the highest share during austral winter. It illustrates the importance of the sources of the Antarctic continent itself, related also to the subsidence of cold air masses also during austral summer, and the importance of synoptic weather patterns like cyclones during austral winter.

These findings clearly demonstrate the value of the inter-disciplinary approach of the CLIMB project, combining expertise in long-term experimental work related to atmospheric chemistry and physics, expertise in Antarctic research campaigns, regional climate modelling, radiative transfer processes, and expertise in state-of-the-art chemical analyses. Although much progress has been made, some gaps have been identified that need to be addressed in future research. In order to discriminate potential source regions even better, more samples would be required, covering several years and with higher temporal resolution, in particular during winter. Measurement sites near the coast have the advantage that these locations are more often impacted by transport from the Southern Ocean and potentially lower latitudes than sites in the Antarctic interior. Simulations of current atmospheric transport models have large uncertainties when simulating many weeks of atmospheric transport what however seems to be necessary. Therefore, more elucidated modelling and input from measurements would be required to disentangle how atmospheric compounds from lower latitudes reach East Antarctica. The regional climate model COSMO-CLM2 was adapted and improved and was found to represent accurately cloud height and occurrence. With respect to cloud ice formation, secondary ice formation, which was not fully implemented in the modified model, is a topic that is mostly unexplored, but which is known to have important effects on cloud structure and cloud phase. Further investigation is also needed to differentiate the effects from the change in cloud phase on the radiative effects from the effects of the change in total water content.

## **Keywords**

Aerosol-clouds-precipitation interaction / Ice nucleating particles / Regional Climate Modelling / East Antarctic Climate / Volatile Organic Compounds

## 1. INTRODUCTION

The water cycle, cloud microphysics and cloud-aerosol-interactions are recognized as key elements of the Antarctic climate system by several international consortia, such as the International Panel on Climate Change (IPCC). Clouds and aerosols play a significant role in the radiative energy budget and aerosols impact cloud microphysics because they are cloud condensation and ice nuclei. In addition, clouds are an important part of the hydrological cycle serving as the agent linking water vapour transport into Antarctica with precipitation. Because precipitation is the only source term in the surface mass balance (SMB) of the Antarctic ice sheet, it is one of the key factors affecting sea level. However, current knowledge on the interaction between clouds, precipitation and aerosol in the Antarctic is still limited, both from direct observations and from regional climate models. This is unfortunate, as the Antarctic ice sheet is expected to become a dominant contributor to sea level rise in the 21st century.

To investigate these complex processes, it is necessary to combine different instrumentation and modelling tools. At the Belgian Antarctic research station Princess Elisabeth (PEA), such an observatory for aerosol, cloud and precipitation properties exists. The synergy of the data sets has been exploited in first case studies on the effect of aerosols on cloud and precipitation processes with an improved aerosol-cloud-precipitation parameterisation of the regional climate model COSMO-CLM2. First results show a strong sensitivity of cloud microphysics to the number of ice nuclei (INP), and to a minor degree to the number of cloud condensation nuclei (CCN). Measurements of INP in Antarctica are, however, rare and challenging because of their very low concentrations. The same is true for VOC measurements in Antarctica. However, VOCs can contribute to atmospheric particle formation and growth and thus are important for the atmospheric budget of CCN and INP.

CLIMB relied on an interdisciplinary research team, bringing together scientists with complementary expertise in long-term experimental work related to atmospheric chemistry and physics, expertise in Antarctic research campaigns, regional climate modelling, radiative transfer processes, and expertise in state-of-the-art chemical analyses.

CLIMB performed measurements of meteorological, aerosol, cloud and precipitation characteristics, both in the vicinity of Princess Elisabeth Antarctica station, and at an altitude which was high enough and exposed to easterly winds to be often within cloud level. In addition to extended ice nuclei particle (INP) filter sampling at PEA, there were (i) a vertically resolved profile of temperature, relative humidity and pressure for three heights: at PEA (1390 m asl), on the Utsteinen nunatak summit (around 1600 m asl) and in the Vikinghogda mountains (2350 m asl); (ii) measurements of precipitation type and intensity by a disdrometer; (iii) measurements of aerosol particle number distribution in the Vikinghogda mountains; and (iv) an automated sampling system for VOCs, also in the remote Vikinghogda mountains. The results were used to improving the aerosol-cloud-precipitation parameterisation in a regional climate model for East Antarctica.



## 2. STATE OF THE ART AND OBJECTIVES

### 2.1 The hydrological cycle over Antarctica

Clouds play an important role in the Antarctic climate and SMB. At first, they are an important component of the Antarctic atmospheric branch of the hydrological cycle, thereby affecting surface mass balance and sea level rise. Secondly, they have an important impact on the surface energy balance. Lastly, a correct representation of the cloud amount and phase is crucial in climate models for the simulation of precipitation timing and the surface energy balance representation.

Unlike in other regions of the world, clouds have a net positive effect on the surface energy balance over polar regions with snow surfaces, both during day- and nighttime (van den Broeke et al., 2006). This is caused by the amount of reflected shortwave radiation being smaller than the amount of backscattered longwave radiation, causing a net positive radiation balance at the surface, i.e., a warming. The amount of backscattered longwave radiation is even larger when liquid or mixed-phase clouds are present, significantly increasing melt and directly affecting the SMB (Van Tricht et al., 2016). Furthermore, the cloud phase is an important parameter determining the net radiative effect at the surface (Matus and L'Ecuyer, 2017). Thin ice clouds can have an important effect on the surface and top-of-atmosphere energy balance in the polar regions (Girard and Blanchet, 2001).

During short measurement campaigns, mixed-phase clouds containing supercooled liquid water at air temperatures as low as -38 to -40°C (below which homogeneous ice nucleation occurs) have been observed both over the Antarctic ice sheet (Lachlan-Cope, 2010) and near the coast (Lachlan-Cope et al., 2016). Identifying ice and liquid-containing clouds is thus of high importance for understanding both precipitation processes and energy balance over the Antarctic ice sheet. Bromwich et al. (2012) provided an extensive overview of existing Antarctic cloud data from various measurement techniques.

Precipitation rates over Antarctica are most often not measured directly. Most studies derive yearly precipitation amounts from ice cores and stake measurements, which are actually records of the local SMB. These records do not consider the removal of precipitation due to (surface and blowing snow) sublimation, which can be considerable over the Antarctic, up to 20 % of the annual precipitation (Déry and Yau, 2002). Furthermore, they neglect disturbances by the transport and redistribution of snow by high wind speeds. Measurements using precipitation radars on the other hand are not affected by these high wind speeds. They provide an estimate of the precipitation intensity by measuring the amplitude of the signal backscattered by hydrometeors. However, to derive precipitation rates from these backscatter values, information about the micro-structure of the snowflakes is necessary, such as their size and shape. Within the AEROCLOUD project ([www.aerocloud.be](http://www.aerocloud.be)), snowfall has been measured independently from accumulation; the latter also including the effect of surface and snowdrift sublimation and redistribution of snow by the wind (Souverijns et al., 2017, 2018a).

The lack of spatial coverage of ground-based precipitation rate measurements can be tackled by using space-borne radars, such as aboard the CloudSat satellite (Wood et al., 2013). However, the satellite has a limited swath width and only overpasses each location of the Antarctic within a range of 100 km on average once every 5 days. This low temporal resolution together with the uncertainty on the microphysical characteristics of the snow particles leads to very high uncertainties in the retrieved precipitation amounts (Souverijns et al., 2018b). Based on these measurements, a mean average precipitation rate of 171 mm year<sup>-1</sup> is obtained over the Antarctic Ice Shield (AIS; north of 82°S; Palerme et al., 2014). Precipitation amounts are however highly variable in different regions, with the interior of Antarctica being considered a cold desert. Apart from a large spatial variability, there is also

a high temporal variability in precipitation amounts (Gorodetskaya et al., 2013) which can be related to the characteristics of the precipitation regime over Antarctica. The annual precipitation amount is dominated by a few high-intensity events (Gorodetskaya et al., 2014) originating from low pressure systems in the Antarctic Circumpolar Trough (Uotila et al., 2011). More and prolonged time series of precipitation and accumulation are therefore highly desirable.

## **2.2 Aerosol observations over 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 itself, 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). Various studies on atmospheric particles have been conducted at different Antarctic research stations during the last decades. A wide range of topics has been investigated, including new particle formation (NPF; Koponen et al., 2003; Asmi et al., 2010; Kyrö et al., 2013; Weller et al., 2015), seasonal cycles of number size distribution and mass concentration (Koponen et al., 2003; Fiebig et al., 2014; Kim et al., 2017), chemical composition (Wagenbach et al., 1988; Teinilä et al., 2000), hygroscopicity (Asmi et al., 2010, Kim et al., 2017; O’Shea et al., 2017) and optical properties of aerosol particles (Weller and Lampert, 2008; Fiebig et al., 2014). In general, there is a yearly trend in particle number concentration, with maximum values in the austral summer. Fiebig et al. (2014) concluded that these cycles are common across the Antarctic plateau, with free tropospheric air masses contributing to air detected at the ground. The highest particle concentrations found in the austral summer are frequently reported to be due to NPF events. Particles formed during NPF events are likely related to sulphate and compounds containing ammonia that were found in the particulate phase in the submicron size range (Wagenbach et al., 1988; Teinilä et al., 2000; Schmale et al., 2013). Precursor gases for NPF can originate from the Southern Ocean (e.g., dimethylsulfide, DMS) and possibly also from other sources, e.g., cyanobacteria in melt water ponds (Kyrö et al., 2013), microbiota from sea ice and the ocean influenced by sea ice (Dall’Osto et al., 2017).

Some studies have reported on Antarctic cloud condensation nuclei (CCN) properties, however, the locations they cover are limited to the Antarctic Peninsula (DeFelice et al., 1997; Kim et al., 2017) or the area of the Wedell Sea on the Brunt Ice Shelf (O’Shea et al., 2017). During the AERO-CLOUD project, measurements of CCN were made at PEA during three subsequent austral summers (2013-2016). Results published in Herenz et al. (2019) clearly showed the presence of background concentrations for both total particle number concentrations and CCN during 61 % of the time. During the remaining time, air masses were observed which had spent time over the Southern Ocean in the 10 days prior

to arriving at PEA. Little is known about ice nucleating particles (INP) in the Antarctic region. Studies with contrasting results exist for the Southern Ocean, where Bigg (1973) obtained much larger INP concentrations than McCluskey (2018). 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. 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 AERO-CLOUD project, Souverijns, 2019a). During a first test in austral summer 2018-2019, filter samples were taken at PEA and analyzed at the Leibniz Institute for tropospheric research (TROPOS, Leipzig, Germany) to obtain atmospheric concentrations of INP. Clearly lower INP concentrations than those for South Pole were obtained. Several new studies have been undertaken in recent years. The Antarctic Circumnavigation Expedition (ACE; Schmale et al., 2019) characterised atmospheric aerosol, their composition and climate effect during a ship-based journey around Antarctica. Tatzelt et al. (2022) analysed with ACE data the abundance of CCN and INP. The Antarctic continent was found to be no important contributor to INP. They found similar INP values like McCluskey (2018) when the air masses had no terrestrial (i.e. non-Antarctic terrestrial) influence and higher INP concentrations when the air masses had terrestrial (i.e. other continents) influences.

### **2.3 VOC observations over Antarctica**

Despite their trace level concentrations, volatile organic compounds (VOCs) play an important role in the atmosphere and influence the global climate. Both anthropogenic activities, as well as biogenic processes, contribute to observed concentrations of VOCs in the troposphere. VOCs are important precursors of ozone and aerosols. Tropospheric ozone is a direct greenhouse gas and plays a key role in the oxidation of atmospheric VOCs, and it is a precursor of hydroxyl radicals in clean environments (Atkinson and Arey, 2003; Griffiths et al., 2021). Typical VOCs such as single-ring aromatic hydrocarbons have an atmospheric life time ranging from several hours to weeks, while there are notable exceptions on both sides of the range. Chlorofluorocarbons (CFCs) atmospheric lifetimes are of the order of decades while e.g. isoprene is transformed with an atmospheric lifetime of 1-2 hours in clean atmospheric conditions (Atkinson and Arey, 2003). Removal of VOCs from the atmosphere happens through the reaction with radical species (e.g. hydroxyl radicals, nitrate radicals, amongst others), photochemical reactions, and reactions with ozone finally resulting in a complete mineralization towards CO<sub>2</sub> and H<sub>2</sub>O. Intermediate products formed during this process result in less volatile compounds with polar functionalities which might form so-called secondary organic aerosols (SOAs) after condensation. Typically, these can also be removed from the atmosphere by wet or dry deposition. Before their removal, SOAs have a substantial effect on the global climate by radiative forcing, both directly via aerosol-radiation interaction responsible for on average -0.22 (-0.47 to -0.04) Wm<sup>-2</sup> and indirectly as they act as CCN, interacting with the cloud formation processes or changing the cloud microphysics, responsible for radiative forcing of on average -0.84 (-1.45 to -0.25) Wm<sup>-2</sup> (Forster et al., 2021; Shrivastava et al., 2017). Next to the effect on Earth's energy budget, clouds play an important role in the hydrological cycle. In Antarctica specifically, precipitation is the only positive contributor to surface mass balance (Boucher et al., 2013; Gorodetskaya et al., 2015).

The atmospheric conditions in the polar regions are very distinct as there are some specific phenomena such as the polar day and night, powerful polar vortices, and strong UV radiation which don't occur anywhere else in the world (Slusher et al., 2010). To understand the influence of VOCs on atmospheric processes, concentration data are of paramount importance. However, in Antarctica, this data remains very scarce. Challenges such as the extreme cold, complicated logistics, and the necessity for robust sampling equipment are the root cause that such a limited amount of studies on these volatile compounds in Antarctica exist. Since the first publication on this topic in 1989, only 22 scientific reports on measurements of VOCs in Antarctica have been published. The first reports describe the sampling and analysis of light nonmethane hydrocarbons (NMHC) (C2-C3) and chlorinated hydrocarbons (CHCs) between 1982 and 1989, using canisters for sampling at Neumayer I station and Scott Base (Rudolph et al., 1992, 1989). Clarkson and Martin (1997) reported a longer time series with shorter intervals for ethane and propane using canister sampling at Scott Base from 1990 to 1996. They observed that mixing of air between mid and high southern latitudes is possible and takes place in a time span comparable to the lifetime of propane (two weeks). Oxygenated VOCs (OVOCs) originating from photochemical oxidation reactions are likely to be present in the Antarctic troposphere as all precursors are present. Only Ciccioli et al. (1996) published data dedicated to OVOCs in Antarctica which were sampled using canisters. A total of 76 different VOCs were identified at 6 sample sites located near Terra Nova Bay during the summer of 1993, proving the ubiquitous occurrence of these compounds.

#### **2.4 Regional Climate Modelling with respect to aerosol-cloud-precipitation interaction**

Besides a lack of observations, there is also only a limited amount of modelling studies. While the results of Global Climate Models (GCMs) are widely available, their output is often lacking in spatial resolution. In addition to that, they are often insufficiently adapted to polar regions, as they frequently use parametrisations that were developed using assumptions that only hold for mid-latitude regions. Regional Climate Models (RCMs) offer the possibility to tackle part of the problems of GCMs. RCMs are limited area models, which dynamically downscale GCM fields. Apart from the higher horizontal resolution and more detailed topography, it is possible to adapt the physics of the RCM for the specific climate over the region of interest. Despite the large role of the AIS in the global climate system, the development of RCMs for the region has been limited, compared to other continents. The regional atmospheric climate model (RACMO2) has been applied for several process- and evaluation-based studies over the Antarctic. This model has been updated throughout the last decades to accurately represent the Antarctic climate and SMB (van Wessem et al., 2018) and has been used to study the past (e.g. Lenaerts et al., 2018) and future of the AIS (e.g. Ligtenberg et al., 2013).

Apart from RACMO2, the Modèle Atmosphérique Régional (MAR) has been applied in several setups over the AIS. Studies of the boundary layer (Gallée et al., 2015) and the SMB (Agosta et al., 2012) have been performed. Furthermore, long-term historical simulations are available (Agosta et al., 2019). A 30-year hindcast for Antarctica has also been created using COSMO-CLM2 (Souverijns et al., 2019b). Another regional climate model that has been tested over Antarctica is MetUM (Walters et al., 2017), while the Danish HIRHAM5 model has been developed with a focus on the northern Polar region (Lucas-Picher et al., 2012; Langen et al., 2017).

To study aerosol-cloud-precipitation interactions, the one-moment schemes for representation of cloud microphysics, which are currently used in many Antarctic models, are unsuitable by design as these models do not relate the drop or ice particle size distribution directly to the aerosol distribution. Comprehensive double-moment cloud microphysics schemes do take into account aerosol types that activate at a given supersaturation (Seifert and Beheng, 2006), and in this way include the aerosol indirect effect. The regional climate model COSMO (CONsortium for Small-scale MOdelling) in its climate mode (COSMO model in Climate Mode, CCLM) does optionally include such a double-moment scheme. In addition, it is a non-hydrostatic model with no scale approximations and applicable especially at the kilometer scale (in our case 2.8 km resolution) and it has been coupled to the Community Land Model under the name COSMO-CLM2. A supplementary aerosol module simulating interactions of atmospheric moisture with Cloud Condensation Nuclei (CCN) and Ice Nucleating Particles (INPs) has been developed by Possner et al., 2017, using the INP parametrisation by Solomon et al., 2015 and the immersion freezing scheme by DeMott et al., 2015.

Within the AERO-CLOUD project model, first tests were conducted using this aerosol scheme for the Princess Elisabeth region (Souverijns et al., 2019a). The models turned out to be sensitive to the number of ice nuclei: When the air is pristine like in Antarctica where few ice nuclei are present, clouds can sustain liquid water layers, which is not the case for higher number concentrations of ice nuclei. Such liquid-containing clouds were also detected by the operational instruments at PEA. Local precipitation is reduced due to the longer residence time of these liquid containing clouds and the cloud radiative forcing is increased due to increased longwave downwelling radiation. The number of ice nuclei appeared to be much more important than the number of cloud condensation nuclei.

## 2.5 Objectives

The objectives of CLIMB were:

- to deliver a unique data set of in-cloud meteorological, aerosol and cloud characteristics, combined with simultaneous boundary layer and ground-based remote sensing measurements;
- to establish a data set with a detailed mapping of air mass origins and transport pathways into East Antarctica;
- to generate an improved COSMO-CLM<sup>2</sup> regional climate model for Antarctica;
- to improve the understanding of the climatological effect of cloud condensation and ice nuclei particles on clouds, precipitation, radiation and the surface mass balance in East Antarctica;
- to strengthen the role of Belgium in the Antarctic research community;
- to valorise the results by scientific publications and workshops, open data access, lectures to the general public and press contributions.

### 3. METHODOLOGY

CLIMB relied on an interdisciplinary research team, bringing together scientists with complementary expertise in long-term experimental work related to atmospheric chemistry and physics, expertise in Antarctic research campaigns, regional climate modelling, radiative transfer processes, and expertise in state-of-the-art chemical analyses. The consortium consisted of two federal research institutes, two Belgian universities and a German research institute.

Princess Elisabeth station (PEA; coordinates: 71.95°S, 23.35°E, 1390 m asl) is excellently located for a study like CLIMB. It is 180 km inland from the coast, in the Sør Rondane Mountains. It is influenced by both synoptic and katabatic meteorological regimes (Gorodetskaya et al. 2013) and access to the Antarctic plateau is fast (half a day on snow mobiles). Therefore, with measurements in the area of PE station, various source regions of air masses and atmospheric particles could be traced back. Note, however, that PEA station is a summer-only station, open from mid-November to end of February, with remote control during the other months. By running over three years, CLIMB covered also inter-annual variability. CLIMB partners are also operating the permanent aerosol, cloud and precipitation observatory (see sections 3.1 and 3.2.1 below), and could use its data therefore in a direct way. CLIMB aimed at performing measurements at an altitude which is high enough and exposed to easterly winds to be often within cloud level. The CLIMB remote site (see Figs. 1 and 2) was located at 2350 m asl, at 72.27101°S and 23.25238°E.

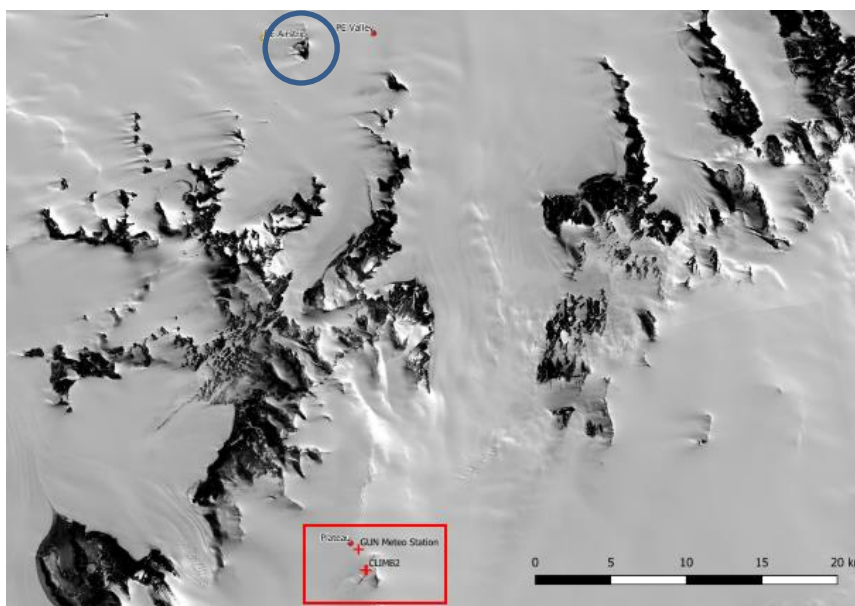


Figure 1: Location of the CLIMB remote site (red box); PEA station in blue circle

CLIMB performed measurements of meteorological, aerosol, cloud and precipitation characteristics in the vicinity of PEA station and directly at the cloud level, with cost- and logistics-efficient small-sized instrumentation. In addition to extended INP filter sampling at PEA, there were (i) a vertically resolved profile of temperature, relative humidity and pressure for three heights: at PEA (1390 m asl), on the Utsteinen nunatak summit (around 1600 m asl) and in the Vikinghogda mountains at the CLIMB remote site (coordinates see above); (ii) measurements of precipitation type and intensity by a disdrometer; (iii) measurements of aerosol particle number distribution at the CLIMB remote site; and

(iv) an automated sampling system for VOCs also at the CLIMB remote site. At the site, an autonomous power system with a wind turbine, solar panels and battery packs were installed (Fig. 2). After two winter seasons (2021 and 2022), everything was dismantled in January 2023.



Figure 2: View of the installation at the CLIMB remote site; left image, from left to right: the pole with the meteorological data logger and the inlet for the optical particle sizer (OPS) attached to it; below the box with the OPS inside and other controlling devices for the VOC sampler and power control, in the middle the pole with the VOC sampler and the solar panel; right image: the wind turbine can be seen and another power-control-battery box under the solar panel

### 3.1 Cloud and precipitation properties

Cloud and precipitation properties were observed at PEA using a ceilometer and a vertically pointing micro-rain radar (MRR), which have originally been installed in 2009. The instruments were kept operational, with major maintenance to the MRR having taken place in the 2021/22 Antarctic season. The ceilometer is measuring cloud heights and optical thickness using a lidar, while the MRR is measuring precipitation properties, mainly intensity and fall speed, using a radar. The database of those measurements has been kept up to date and the instruments' operation has been supervised remotely, including during the unmanned periods. However, power outages, especially in the austral winter months, limit the continuity of the instruments' operation during those times of the year. In the last austral winter, we therefore increased the frequency of controls for both instruments in order to ensure that they operate normally and are restarted after a power outage, resulting in a more consistent dataset.

Observations are first stored locally on a server at the station, from where they are transferred via a satellite connection to servers in Amsterdam, before they are post-processed, stored and backed up at KU Leuven. From the ceilometer data, as part of a bachelor thesis, we identified cloudy versus clear sky periods using the method proposed by Van Tricht et al. (2014), as well as the cloud phase according to the work by Guyot et al. (2022). The thresholds in both methods were slightly adapted to the local circumstances. The resulting periods of occurrence of the different cloud phases can then be analysed with respect to their duration and weather conditions under which they occur.

## 3.2 Characterisation of physical properties of atmospheric aerosol

### 3.2.1 Physical aerosol properties within boundary layer at PEA, remote site and for INP

#### 3.2.1.1 Physical aerosol properties within boundary layer at PEA

Three instruments of the permanent aerosol observatory were operational during the reporting period (located in the 'Southern Scientific Shelter' of PEA): the TEOM-FDMS (particle total mass concentration; Thermo Scientific), the nephelometer (particle light scattering coefficient; Ecotech Aurora 3000) and the aethalometer (particle absorption coefficient and equivalent Black Carbon concentration; Magee Sci. AE-31). The aethalometer was shipped in November 2020 after repair in Belgium and re-started operation in the beginning of December 2020. Beginning of June 2021 to November 2021 there was a data gap due to a power outage of PEA. The TEOM-FDMS could not be restarted in November 2021 and had to be shipped back for repair. It was re-installed in December 2022. Two other instruments (condensation particle counter and Laser Aerosol Spectrometer) were in Belgium for repair and re-installation at PEA is foreseen for season 2023/2024.

#### 3.2.1.2 Aerosol properties at CLIMB remote site

For measuring the particle number size distribution (PNSD), an optical particle sizer (OPS, TSI Inc., model 3330) was used. It measures between 300 nm and 10  $\mu\text{m}$  particle diameter. As particles which are active as cloud condensation nuclei (CCN) or ice nuclei (INP) are mostly  $> 500$  nm (e.g. DeMott et al., 2015), this type of device can deliver good information to estimate the general availability of CCN or INP. Unfortunately, the OPS could not be purchased in time for the field season 2020/21 due to COVID-19 pandemic related reasons. It has been tested during 2021 and finally installed at the CLIMB remote site in January 2022 (Figs. 2 and 3). It was connected to a mini-pc in order to restart operation after potential power cuts. However, after ten days of normal operation at the remote site, the flow got apparently blocked and measurements stopped (instrument power for the OPS stopped however only 2 months later).



Figure 3: inside view of the box at the CLIMB remote site with OPS and other controlling devices



### 3.2.1.3 Ice Nuclei Particle Sampling

The sampling for the INP filter measurements was set up in a shelter around 500 m north of PEA station ('Atmos shelter'). The filter holder for the 47 mm PTFE filters was situated outside, on top of the shelter with a 15 cm long piece of flexible conductive tubing as inlet (see Fig. 4). Pump and flowmeter were inside the shelter. The sample volume flow was around 22 – 24 L/min. Temperature and pressure for calculation of ambient and standard conditions were taken from the automatic weather station (AWS) of PEA and from the flowmeter connected to the sampling line, respectively. Sample duration was around 10 days per filter. Each season, three to four blank samples were taken. During the campaign 2020/21, a total of 6 samples, plus 3 blank samples, could be collected. Another set of 6 samples, plus 3 blank samples, could be collected between end of November 2021 and mid-February 2022.

The INP samples were analysed by CLIMB subcontractor TROPOS (Leibniz institute for tropospheric research, Leipzig, Germany). Two different methods, called LINA (Leipzig Ice Nucleation Array) and INDA (Ice Nucleation Droplet Array) were used for the filter evaluation (Wex et al., 2019; Hartmann et al., 2019). The filters were washed with ultrapure water and were examined in the freezing arrays, as follows: LINA is a plate cooled by a Peltier element onto which 90 droplets of the washing water with a volume of 1  $\mu\text{L}$  each are placed which are then observed with a camera while the plate is cooled. The second method uses polymerase chain reaction (PCR)-plates with 96 wells, and a droplet of 50  $\mu\text{L}$  is placed in each. The PCR-plate is sealed and immersed in a thermostat, and droplets are also monitored during the cooling process. For more detail, see e.g., Gong et al. (2019). The parameter that is obtained is a temperature dependent fraction of frozen droplets which can be converted to a concentration of INP in the atmosphere.



Figure 4: inlet setup for the INP sampling on the roof of the northern 'Atmos' shelter

### 3.2.2 Aerosol properties at PEA derived from remote sensing and from radiative transport modelling

BIRA operated an Airyx 2D MAX-DOAS and a CIMEL sunphotometer from the roof of PEA (Fig 5). The MAX-DOAS was installed in November 2019 and it ran discontinuously until 12 December 2022, when control of the instrument was lost. The MAX-DOAS is based on two spectrometers operating in the UV and Visible spectral range, and an optical head able to scan the full hemisphere to collect the light and send it through an optical fiber to the spectrometers. The main direction of measurements was pointing north ( $17^{\circ}\text{E}$ ), although we pointed south during the first months. The MAX-DOAS performed elevation scans between zenith and the horizon at a fixed azimuth (mainly  $17^{\circ}\text{E}$ ) and almucantar scans, i.e. measurements at the elevation of the sun and in different relative azimuths. As described below, we used elevation scans and zenith measurements to respectively retrieve aerosol optical depths (AODs) and stratospheric abundances of ozone,  $\text{NO}_2$ , and OClO. Table 1 shows the operations of the MAX-DOAS during the CLIMB project.

We installed the CIMEL at the beginning of each season and it came back each year for calibration. The CIMEL measures the absolute light intensity at several wavelengths. The CIMEL points to the sun to retrieve AODs, it also performs almucantar and principal plane scans, i.e. measurements at various elevations at the azimuth of the sun.

Table 1: Operations of the MAX-DOAS during CLIMB project

30 November 2020-6 December 2020	Installation and nominal
6 December 2020 – 23 February 2021	Pointing south, reduced surface sensitivity
23 February 2021 – 1 June 2021	Nominal
1 June 2021 – 16 December 2021	Power cut and ice on the tube
16 December – 24 April 2022	Nominal
24 April 2022 – 20 August 2022	Polar night, no MAX-DOAS data
20 August 2022 – 13 November 2022	Nominal
13 November 2022 – 26 November 2022	Elevation motor blocked
26 November 2022 - 12 December 2022	Nominal

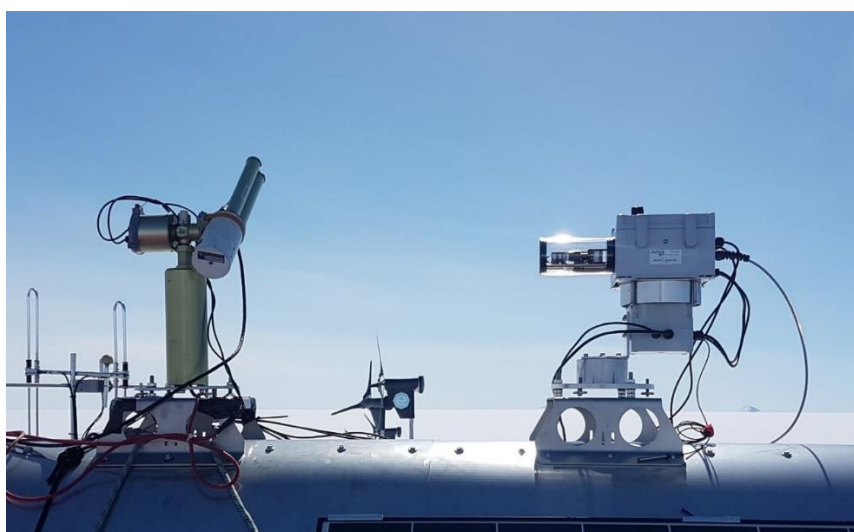


Figure 5: The CIMEL sunphotometer (Left) and Airyx MAX-DOAS (Right) on the roof of the PEA station.

The spectra measured with the MAX-DOAS instrument pointing towards different elevation angles were successively analyzed with the spectral analysis tool QDOAS (Dankaert et al., 2013) and the profiling tool MMF (Friedrich et al. 2019) applying the MAX-DOAS technique (e.g. Hönninger et al., 2004, Platt and Stutz, 2008). The retrieved aerosol optical depths (AODs) were then compared with those estimated by the CIMEL-sun-photometer described earlier.

In the first step, differential slant column densities (dSCDs) of different molecules were fitted to the spectra in two wavelength windows (450 nm -- 540 nm and 338 nm -- 370 nm, visible and UV, respectively) in order to estimate the dSCD of the oxygen dimer (O<sub>4</sub>) at wavelengths close to the center of the wavelength windows, located at cross section peaks of O<sub>4</sub> (360 nm and 477 nm). Cross section references and other QDOAS settings for both spectral fitting windows are summarized in Table 2.

Table 2: Settings for the spectral analysis software QDOAS used for the O<sub>4</sub> dSCD retrieval in the two fitting windows.

425–490 nm	
O <sub>3</sub> (223K)	Serdyuchenko et al. [2014]
Ring Effect	Chance and Kurucz [2010]
O <sub>4</sub> (293K and 203K)	Thalman and Volkamer[2013]
H <sub>2</sub> O(293K)	HITEMP [Rothman et al.,2010]
NO <sub>2</sub> (273K and 223K)	Bogumil et al. [2000]
Intensity offset	Constant
Reference	Daily noon
338-370 nm	
BrO (223K)	Fleischmann et al. [2004]
O <sub>4</sub> (293K)	Thalman and Volkamer [2013]
O <sub>3</sub> (243K and 223K)	Serdyuchenko et al. [2014]
HCHO (297K)	Meller and Moortgat [2000]
Ring Effect	Chance and Kurucz [2010]
NO <sub>2</sub> (273K and 223K)	Bogumil et al. [2000]
Intensity offset	Constant
Reference	Daily noon

In a second step, the dSCDs estimated at wavelength 360 nm and 477 nm were used as input to the profiling algorithm tool MMF, in order to retrieve profiles of the aerosol extinction coefficient and total AODs. The profiling tool MMF is based on the constrained least-square fitting method applying the Levenberg-Marquadt iteration scheme (Levenberg, 1944) and it uses a regularization matrix constructed from an a priori profile assuming an exponentially decreasing extinction profile with a scaling height of 1 km and a fixed AOD of 0.04. The constructed covariance matrix assumes 50% variation on the diagonal and a correlation length of 250 m to estimate the off-diagonal elements. The forward model step of MMF exploits the radiative transfer model VLIDORT v.2.7 (Spurr et al., 2001, 2006, 2013).

In the framework of the ESA FRM4DOAS project (<http://frm4doas.aeronomie.be/>), a Round Robin exercise of profiling algorithms was carried out, both on synthetic data (Friess et al., 2019) with medium and high aerosol loads and cloud layers, as well as on data gathered during the CINDI-2 campaign (Tirpitz et al., 2021), which showed a good performance of MMF for aerosol retrievals under aerosol free, and medium (AOD=0.1) to high (AOD=1.0) aerosol load conditions as well as under cloudy conditions. While MMF can operate in the linear as well as in the logarithmic retrieval space, this study was only performed with the logarithmic setting since studies for the above mentioned Round Robin exercise showed better results for MMF when working in logarithmic retrieval space.

Previous studies (e.g. Wagner et al. 2019) showed a potential need for a scaling factor of the O4 dSCD, and analysis with a complementary profiling algorithm during routine FRM4DOAS retrieval with MAPA (Beirle et al. 2019) which allows the scaling factor to be fitted, show an average scaling factor between 0.8 and 0.9 for the UV range and values above 0.9 for the VIS range. We therefore decided to include such a scaling factor in the analysis of the dSCDs at Utsteinen, we choose a scaling factor of 0.9 as the default for the UV and 0.95 for the VIS.

The summarized settings for MMF are:

- temperature and pressure profiles: from a climatology (used within the FRM4DOAS framework) created from 20 year averages of ECMWF reanalysis from 1995 to 2016 provided by Steffen Ziegler, MPIC.
- A-priori profile: exponentially decreasing with AOD=0.04 and a scaling height of 1 km
- A priori covariance matrix Sa: 50% of the a priori profile on the diagonal, Gaussian dependence for off-diagonal terms with a correlation length of 250 m
- Surface albedo: 0.9
- down-scaling factor of measured dSCD before processing: 0.9 (UV), 0.95 (VIS)
- Henyey Greenstein phase function with an asymmetry factor  $g=0.7$
- Single scattering albedo: 0.98 (typical values from RMI nephelometer and aethalometer)
- Retrieval altitude grid: 20 layers of 0.2 km thickness from 1.39 km to 5.39 km a.s.l. and a simulation grid reaching up to 60 km.
- Quality control criteria based on the degrees of freedom (DOFS) and the RMS of the difference between measured and calculated O 4 dSCDs

### 3.3 Characterisation of (semi-)volatile organic compounds

#### 3.3.1 Development of the autosampler

A new automated sequential sampler (ATS-50) was developed and optimized in-house within the EnVOC research group of Ghent University, Belgium, to sample VOCs autonomously over a period of one year via active sampling on stainless steel sampling enrichment tubes filled with 200 mg Tenax TA as a sorbent. The developed sampler uses a rotating manifold and stepper motor to select the sampling positions (Figure 6) and was controlled using an open-source Arduino microcontroller. During the CLIMB project several upgrades were implemented to cope with the extreme cold such as

a stronger motor, PTFE sleeved O-rings, closed loop motor control with an encoder) custom mechanical sensor to find 0 position, rigidity upgrades.

Passive uptake of VOCs by diffusion whilst the tubes are inserted into the autosampler is minimized by the use of sorbent tubes outfitted with a diffusion limiting insert (SafeLok) increasing the diffusion path length (Markes international, 2014; Woolfenden, 2012; Woolfenden and Cole, 2003). The autosampler is designed to be equipped with up to 50 sorbent tubes and 5 blanks which are held in an identical configuration, but not sampled. The main body of the sampler consists of food-safe polyacetal polymer (POM) and stainless steel. All tubing and connections used are made of either PTFE or stainless steel. Developing our own autosampler was essential to achieve the necessary flexibility in the programming of our sampling campaign, and to sample under extreme conditions posed by the high wind speeds and cold temperatures. Mechanical and electrical components which remain stable at low temperatures were selected for its construction. An accurate sample volume is crucial for the final calculation of the VOCs concentration. Therefore, a Honeywell Zephyr HAF amplified flowmeter (0-200 sccm) was inserted between the sample tubes and the pump (Thomas G12/02EB, oil-less rotary vane, Germany). Flowrate and sampling time were logged continuously on an industrial (-40°C) SD card (Innodisk 512 MB). To validate the flowrate, a calibrated TSI 4140 flowmeter was used. The autosampler was programmed at a flow rate of 30 sccm for 3 days resulting in a total planned sample volume of 130 L on 1 sorbent tube.

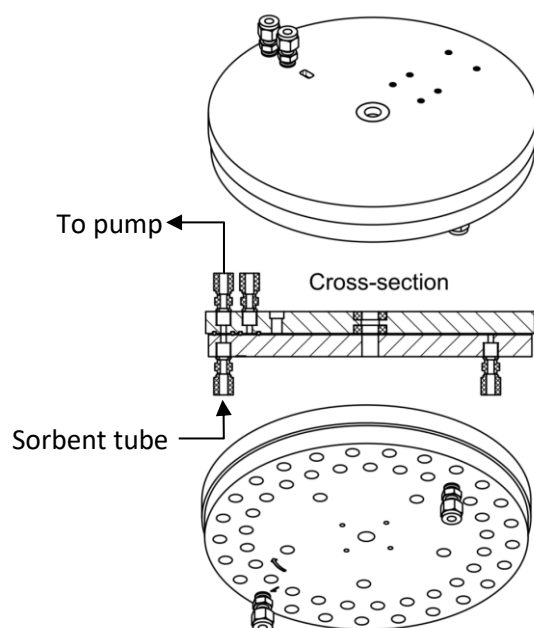


Figure 6: schematic of the autosampler

During the 2019-2020 sampling 1 autosampler was used with 25 samples near the station. During the 2<sup>nd</sup> campaign (2020-2021) the sampler near the station was reused and one new sampler incorporating some design changes was used at the CLIMB remote site, both filled with 25 samples. During the last field campaign (2021-2022), only the latest version of the autosampler was used at the CLIMB remote site filled with 50 samples.

### 3.3.2 Analysis of (O)VOCs by TD-GC-MS

The collected samples were analyzed following two discrete approaches. Firstly, the samples were analyzed by TD-GC-MS. The split flow, containing 75% of the original sample material, was recollected on a conditioned sorbent tube (200 mg Tenax TA). The recollection tube was then analyzed by an innovative and in-house developed TD-PTR-Qi-TOFMS combination which couples a thermal desorber to a high-resolution PTR-MS (see also Figure 7).

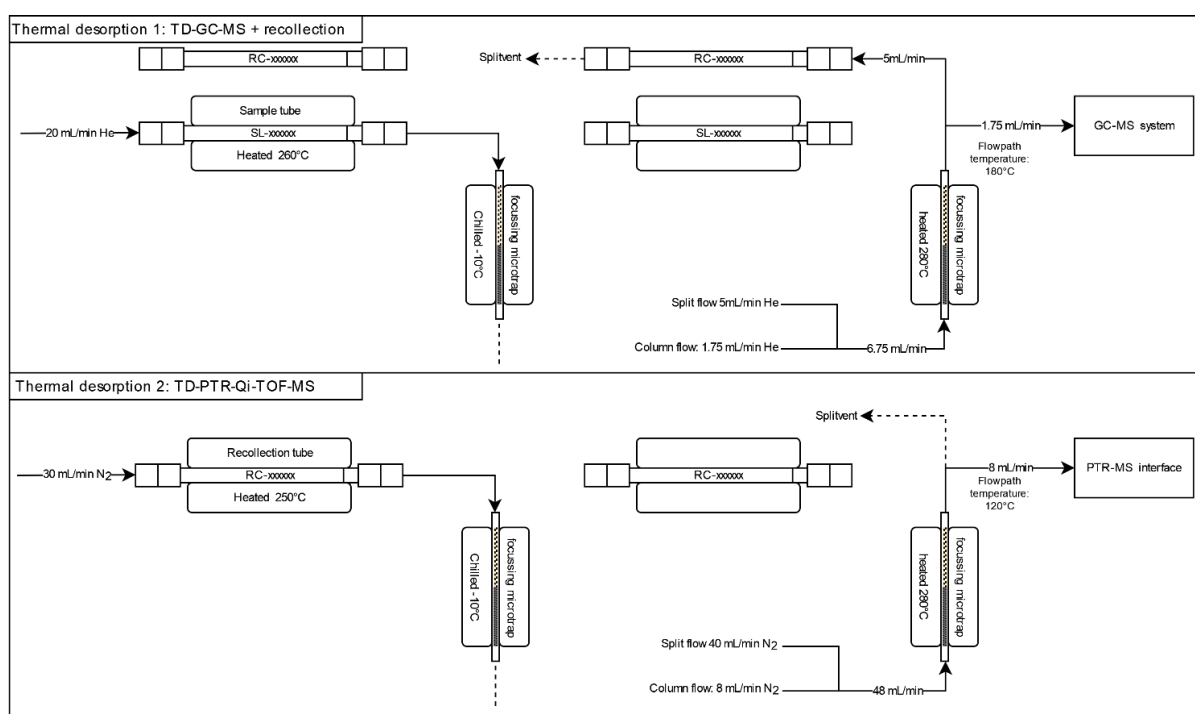


Figure 7: Analysis sequence used to desorb thermal desorption samples in 2 instruments

Samples were analyzed on a 2-stage thermal desorption GC-MS system. Thermal desorption was performed using a Markes (Llantrisant, UK) series 2 Unity coupled with an Ultra autosampler. The analytes were refocused on a microtrap before injection on the GC-column. Electron ionization was used (70 eV) as an ion source for the quadrupole mass selective detector.

### 3.3.3 Analysis of (O)VOCs by TD-PTR-Qi-TOFMS

A high-resolution PTR-Qi-TOFMS instrument (Ionicon, Austria) was hyphenated to a thermal desorption system consisting of a Unity and an UltraTD (Markes, Llantrisant, UK). This home-made setup allows for direct desorption of the sorbent tube containing the recollected sample (see section 3.3.1 and Figure 8 **Error! Reference source not found.**) into the PTR-Qi-TOFMS.

The analytes are ionized by chemical ionization (using  $\text{H}_3\text{O}^+$ ) and subsequently analyzed by high-resolution time-of-flight mass spectrometry. The interfacing between the two instruments is schematically depicted in **Error! Reference source not found.**. A stainless-steel tee union (1/16", Swagelok) was used to dilute the gas flow (8 sccm) coming from the thermal desorption unit with nitrogen (diluent gas) to compensate for the higher inlet flow (80 - 180 sccm) of the PTR-Qi-TOFMS

instrument. The Unity transfer line was inserted 20 mm inside of the PEEK tubing which was connected to the PTR-Qi-TOFMS inlet. This avoids contact of the analytes with the metal surface.

This setup is only rarely used but yields very interesting results due to the sensitivity of the PTR-MS to oxygenated VOCs. To our knowledge this is the first time the same sample is analysed sequentially by TD-GC-MS and PTR-MS, which creates new analytical possibilities for a broad range of applications.

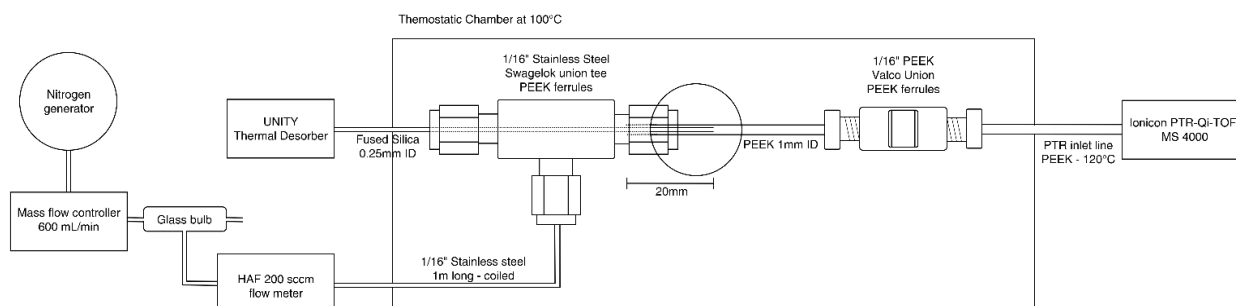


Figure 8: Interface between the thermal desorber and the PTR-Qi-TOFMS developed for the analysis of OVOCs in the air samples from Antarctica

### 3.4 Meteorological analysis

#### 3.4.1 Meteorological analysis from disdrometer, mini-sensors, radio soundings and AWS

The disdrometer (Lufft WS100; Germany) for detecting precipitating and type (snow or drops) could not be purchased in time for the field season 2020/21, like the OPS and due to COVID-19 related issues. Finally, the disdrometer has been shipped to PEA for field season 2021/22, tested there and finally installed end of December 2021 on the southern roof of PEA, near to the micro-rain radar and ceilometer (Fig. 9). Since then it is operating continuously. Unlike the micro-rain radar, it does not give vertical information. It detects only precipitation that reaches the disdrometer.

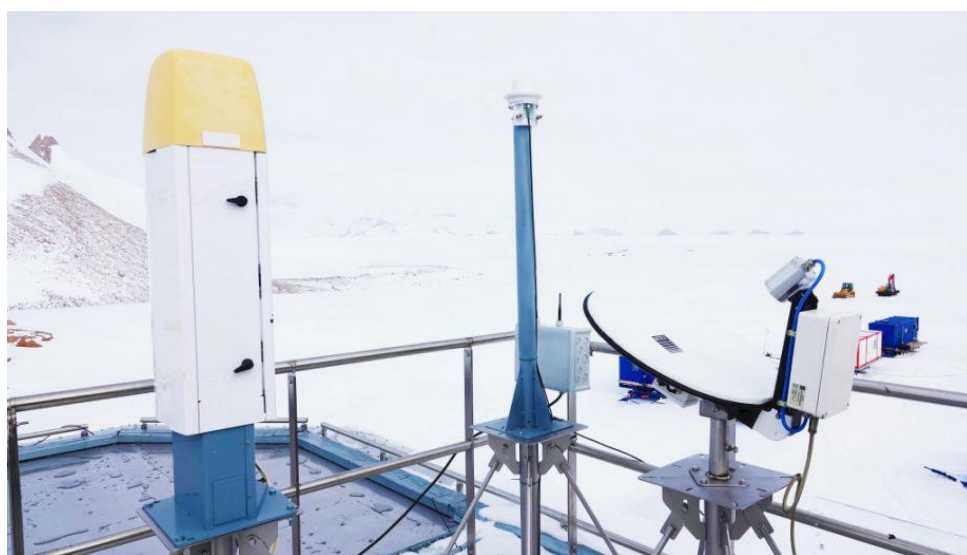


Figure 9: the ceilometer (left), disdrometer (middle) and micro-rain radar (right) on the southern roof of PEA

Likewise, the robust small meteodata loggers (Driesen+Kern, DK323 HumiBaroLog, Germany) could not be purchased in time for field season 2020/21. Three sensors (each for temperature, relative humidity, atmospheric pressure) were tested at RMI before shipment for field season 2021/22. On the one hand they were measuring next to each other over several weeks (intra-comparison), and on the other hand all three measured, over one week, together with standard sensors in the calibration laboratory of RMI (cycles of very low to high temperatures, pressure adapted to altitudes of the final installation sites, various levels of relative humidity). The three meteo data loggers have been installed as foreseen: one at PEA, on the roof of the southern scientific shelter; one on the Utsteinen nunatak, and one at the Climb remote site (see Figs. 2 and 10). The logger at the remote site collected data until end of April 2022, the one on the nunatak without interruption, and the logger on the southern science shelter until beginning of June 2022. During field season 2022/23, the batteries of the loggers were exchanged, and the loggers restarted and re-installed: the ones from the southern science shelter and the nunatak continue operation at these locations, and the logger from the CLIMB remote site has been installed at the northern ‘Atmos’ shelter of PEA.



Figure 10: the meteorological data loggers on the Utsteinen nunatak (left) and on the southern science shelter (right)

The automatic weather station (AWS) of IMAU, Utrecht, installed 500 m to the East of PEA, had to be de-installed because of empty power supplies and end of lifetime of some components. It stopped recording already in July 2020. The de-installation went without issues and the AWS was sent back to Belgium and finally to IMAU in March 2021. The data has been processed by IMAU and is available via partners RMI or KUL. Alternative, current AWS data is available from the AWS near the Utsteinen air strip (IPF) and from the AWS east of Utsteinen ridge (EPFL, Lausanne, Switzerland). Data from the AWS near the air strip is available up to mid-February 2022.



Radio soundings by weather balloons were performed during season 2020/2021, 2021/22 and 2022/23, approximately each second day and data have been sent to the Global Telecommunication System (GTS). The radio sounding data have been analysed with respect to, e.g., tropopause altitude, inversion layers, statistics of temperature, relative humidity, wind, and integrated water vapour.

#### 3.4.2 Back trajectory and dispersion modelling

Air mass tracing has been done by dispersion analyses of atmospheric trajectories, using the atmospheric dispersion model FLEXPART, in order to determine the most probable source and receptor regions and atmospheric pathways. FLEXPART (Stohl et al, 2005) 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. The model can also be used in a forward or backward mode, making it possible to trace back the source contribution of a certain air component (Stohl et al, 2012, De Meutter et al., 2018). FLEXPART was driven by meteorological input data from the European Centre for Medium-Range Weather Forecasts, using the ERA-5 data set at a grid of  $0.5^\circ \times 0.5^\circ$ . The dispersion analysis yielded probability functions for the source regions and atmospheric pathways.

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, for the whole period and also for each season separately. When the clustering is performed relying on the normalised latitude, longitude and altitude, four clusters of air mass origin are found. In addition, the backward trajectories have been combined with distinct parameters like particle number concentration, aerosol absorption exponent, potential vorticity, exposure to sunshine duration. 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 2017/2018 up to 2020/2021.

### 3.5 Regional Climate Modelling

We developed a modified version of COSMO-CLM<sup>2</sup> (Souverijns et al., 2019b; Possner et al., 2017), which implements a two-moment scheme (Seifert and Beheng, 2006) and a parametrization for aerosols and their interaction with atmospheric water, including INP and CCN. This model allows for a prescription of concentrations for those aerosol types, which permits an investigation of the influence of said aerosols on atmospheric properties such as cloud water phases, and how well the parametrisation of the aerosols matches with the observations of cloud properties and aerosol concentrations by the other part of the project. Furthermore, the model features a radiative scheme which was used to examine the effect of the different cloud structures on cloud radiative properties.

While initially it was planned to integrate the model for a period of ten years, the simulation length had to be reduced due to computational limitations. We tested integrating the model using the output of a coarser integration of the faster standard version of COSMO-CLM<sup>2</sup> as boundary conditions, which would have allowed us to reduce the spatial extent of the simulated domain, and thus would have meant an increase in computation speed. However, we found that a larger spatial extent in

combination with using ERA-5 data as boundary conditions resulted in a much better representation of observed cloud structures. Furthermore, as early tests showed a negligible influence of CCN, we restricted ourselves to investigating the influence of INPs. For those, we selected five settings (Table 3), with the first corresponding to the low end of INP concentrations we observed at the station, and the second corresponding to the high end of INP concentrations we observed at the station. The third setting corresponds to the low end of mid-latitude continental or the high end of maritime INP observations. In addition to that, we used this third setting to compensate for the missing secondary ice processes of the model in the second setting, as those would increase the ice crystal number concentration above the level obtained from our model. The fourth and fifth settings then correspond to what can be seen as an average mid-latitude continental value and a high-end mid-latitude continental value, which we used to compare the results from the other settings to.

Table 3: Overview of the prescribed INP concentrations

INPs at -20°C	Representative of	Comment
$1 \cdot 10^{-5} \text{ L}^{-1}$	Low-end Antarctic	
$5 \cdot 10^{-3} \text{ L}^{-1}$	High-end Antarctic	
$5 \cdot 10^{-2} \text{ L}^{-1}$	Average Maritime; Low-end Mid-Latitude continental; High-end Antarctic accounting for secondary ice	
$2 \cdot 10^{-1} \text{ L}^{-1}$	Average Mid-Latitude continental	Austral summer only
$2 \text{ L}^{-1}$	High-end Mid-Latitude continental	

## 4. SCIENTIFIC RESULTS, IMPLICATIONS AND RECOMMENDATIONS

### 4.1 Cloud and precipitation properties

#### 4.1.1 Results from the ceilometer and micro-rain radar

An overview of the compiled data obtained from the ceilometer and the MRR can be found in Table 4. The resulting datasets are available to the scientific community upon request, and we are exploring other possibilities of making the data available. As such, we shared the ceilometer data with Bodeker Scientific, a research organization from New Zealand (<https://www.bodekerscientific.com/>), who are compiling a dataset of cloud observations from Antarctica. Preliminary results of the cloud phase analysis, where liquid water was identified using the algorithm by Guyot et al. (2022), can be seen in Figure 11. It is evident that liquid-containing clouds were much more frequent during austral summer, when in about a third of all clouds, liquid water has been detected. These events were found to correlate to more northerly winds in the upper atmosphere.

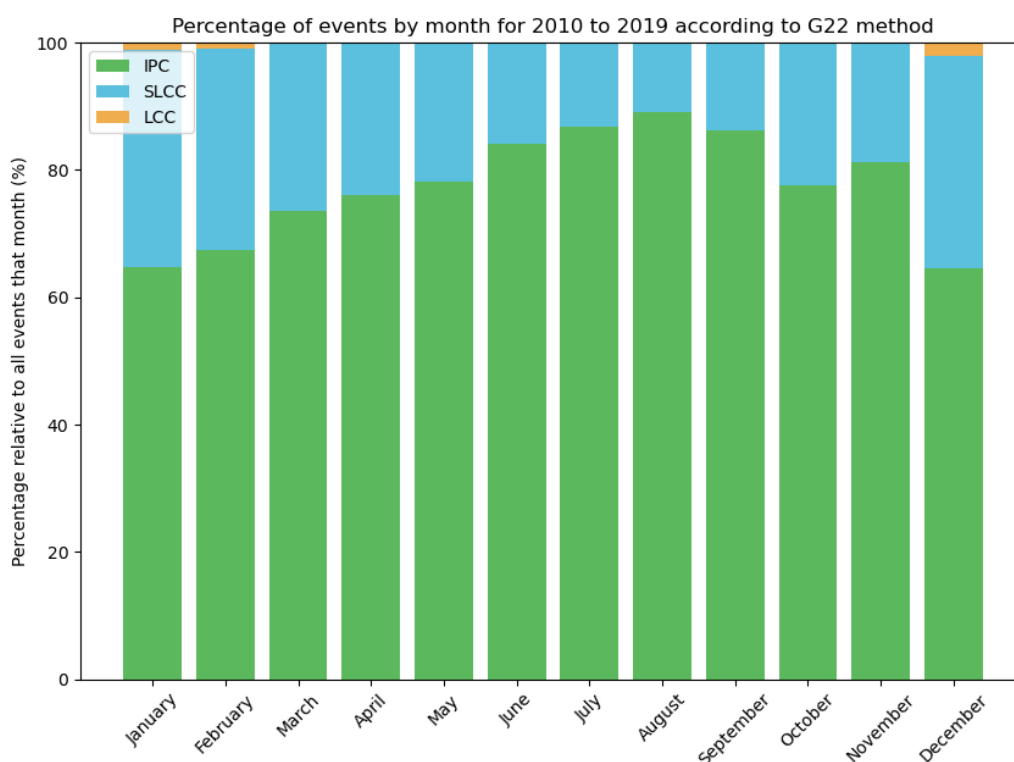


Figure 11: Percentage of (supercooled) liquid containing clouds per month, out of all clouds that month, derived using the method by Guyot et al. (2022), summed over all Ceilometer measurements 2010-2019. IPC = Ice and Precipitating Clouds, SLCC = Supercooled Liquid Containing Clouds, LCC = Liquid Containing Clouds.

Table 4: Overview of available recordings for the MRR and the Ceilometer. F = fully available (<3 days missing), P = partially available (>3 days missing), N = no measurements available.

	Jan	Feb	Mar	Apr	Mai	Jun	Jul	Aug	Sep	Oct	Nov	Dec	
2010	P	F	P	P	N	N	N	N	N	N	N	P	MRR
	P	P	P	N	N	N	N	N	N	N	N	N	Ceilometer
2011	F	F	F	P	N	N	N	N	N	N	N	P	MRR
	P	F	F	P	N	N	N	N	N	N	N	P	Ceilometer
2012	F	F	P	F	F	F	F	F	F	F	F	F	MRR
	F	F	P	N	N	N	N	N	N	N	N	N	Ceilometer
2013	P	F	P	P	P	N	N	N	N	N	N	P	MRR
	F	F	F	F	P	N	N	N	N	N	P	P	Ceilometer
2014	P	P	F	P	F	F	N	N	N	N	P	F	MRR
	N	P	F	F	P	N	N	N	N	N	N	P	Ceilometer
2015	F	F	F	F	F	P	N	N	N	P	F	P	MRR
	P	F	F	P	F	P	P	F	F	F	P	P	Ceilometer
2016	F	F	F	F	P	N	N	N	N	N	N	N	MRR
	F	F	F	F	P	N	N	N	N	N	P	P	Ceilometer
2017	N	N	N	N	N	N	N	N	N	N	N	P	MRR
	F	P	N	N	N	N	N	N	N	N	N	P	Ceilometer
2018	P	P	F	P	N	N	N	N	N	N	N	P	MRR
	P	F	F	F	N	N	N	N	N	N	P	P	Ceilometer
2019	P	F	P	N	N	N	N	N	N	N	N	P	MRR
	F	P	P	N	N	N	N	N	N	N	P	P	Ceilometer
2020	F	P	N	N	N	N	N	N	N	P	P	P	MRR
	P	F	F	F	F	F	F	F	F	P	P	F	Ceilometer
2021	P	F	N	N	P	P	N	N	N	N	P	F	MRR
	F	F	F	F	P	N	N	N	N	N	P	F	Ceilometer
2022	F	P	N	N	N	N	P	F	F	F	F	F	MRR
	F	F	F	F	F	F	F	F	F	F	F	F	Ceilometer
2023	F	F	F	F									MRR
	F	F	F	F									Ceilometer

## 4.1.2 Implications and Recommendations

While there are now other locations in Antarctica with similar instrumentation, none of those have been running for as long as the observatory at PEA. With the recent maintenance, we have improved the availability of measurements, and we hope to expand the dataset in the future. The measurements are an important resource for climate modellers, as it can be used and is being used to verify model output in an area where observations are very sparse, with an observation frequency that is impossible to reach using satellites. In particular, the occurrence and the base height of clouds can be verified using the ceilometer data, as well as, through further processing, the occurrence of liquid water. Snowfall rates can be verified using the MRR.

In the upcoming PAsPARTOUT project, it is planned to correlate cloud properties with new, improved aerosol samples. As such, it is necessary to continue the measurements. On the long-term, it may become necessary to replace or upgrade the measurement hardware when they reach the end of their lifespan. The manufacturer of the MRR advises to upgrade to the newest generation of the instrument. Finally, the datasets have been examined in various Bachelor and Master theses, where it provided students with the chance to improve their data-processing skills on a first-hand, real-world scientific dataset, which is something we would like to continue.

## 4.2 Characterisation of physical properties of atmospheric aerosol

### 4.2.1 Physical aerosol properties within boundary layer at PEA and at remote site

From the long-term aerosol measurements at PEA, it was found that the overall amount of particles in the Antarctic atmosphere is very low (by a factor of around 10-100 lower than in Europe). 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. Further, Figure 12 illustrates the particle number in the size ranges 300 to 500 nm, 500 to 1000 nm and larger than 1000 nm, measured at PEA and showing that larger particles are only present in low concentrations. This indicates a strong scarcity of available particles for cloud condensation nuclei (CCN) and ice nuclei (INP) because such particles are mostly of sizes larger than 500 nm.

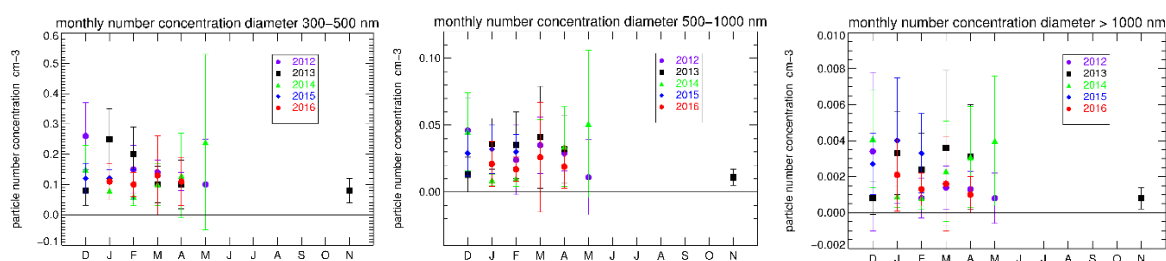


Figure 12: particle number concentrations in the size range 300 to 500 nm (left), in the size range 500 to 1000 nm (middle), and in the size range larger than 1000 nm (right, up to 7000 nm); measured at PEA.

The optical particle sizer (OPS) was first installed in the southern scientific shelter of PEA for testing purposes. The measured values agreed well with the respective size distribution measured with the Laser Aerosol Spectrophotometer at PEA. Concentrations for larger particles decreased distinctly to values  $< 0.05$  particles/cm<sup>3</sup>. Finally, the OPS could measure for around 10 days at the CLIMB remote site (Fig. 13). It can be seen that particle numbers in the different size ranges were in line with the values measured at PEA. However, they tended to be lower, in particular with respect to particle numbers larger than 1000 nm.

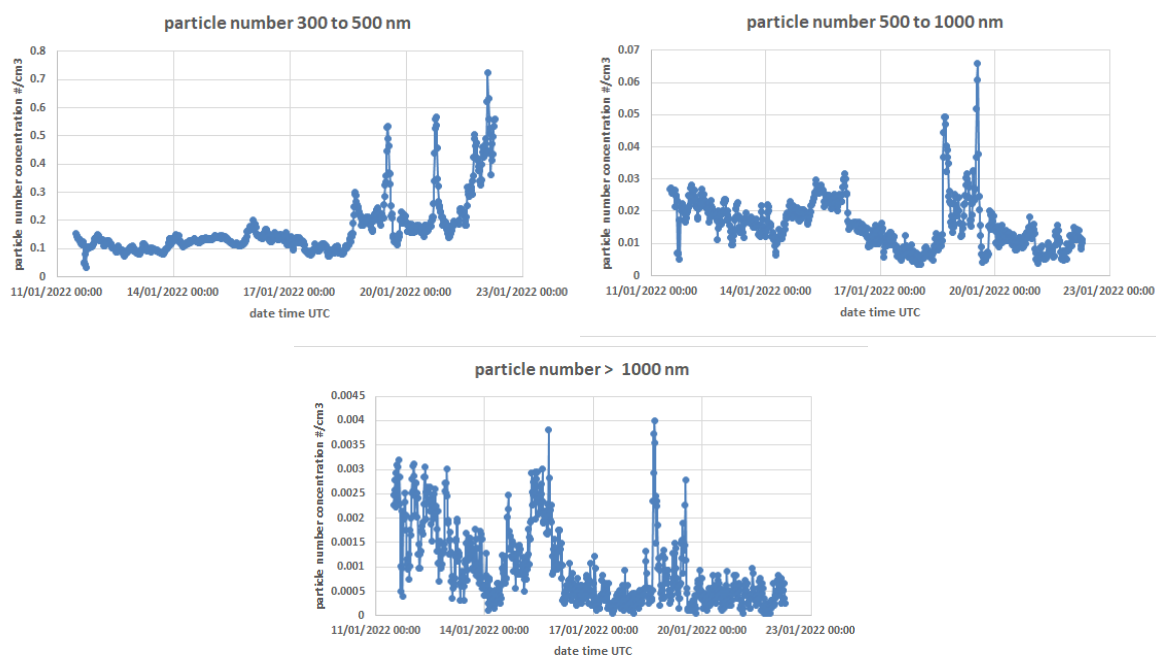


Figure 13: particle number concentrations in the size range 300 to 500 nm (upper, left), in the size range 500 to 1000 nm (upper, right), and in the size range larger than 1000 nm (bottom, up to 10 000 nm); measured at PEA.

The peaks in number concentration on 18 and 19 January, visible within all size ranges, occurred during stormy weather conditions with precipitation. The size range 300 to 500 nm showed afterwards an increase in number, but not the other size ranges which showed a decrease in number. There was no precipitation during those later days but still high wind speeds. This indicates that the air masses after the stormy weather conditions were depleted of larger particles, but apparently enriched by particles of 300 to 500 nm of size.

#### 4.2.2 Ice nucleating particles at PEA

Figure 14 shows results of the analysed INP filters taken at PEA during the field seasons 2018/19 and 2020/21. The samples of season 2021/22 are preliminary analysed, but not all necessary corrections are done. At PEA, low INP concentrations were obtained. Compared to INP samples collected at the German Antarctic station Neumayer III, and also compared to the scarce literature data, the INP numbers for PEA are at the lower limit.

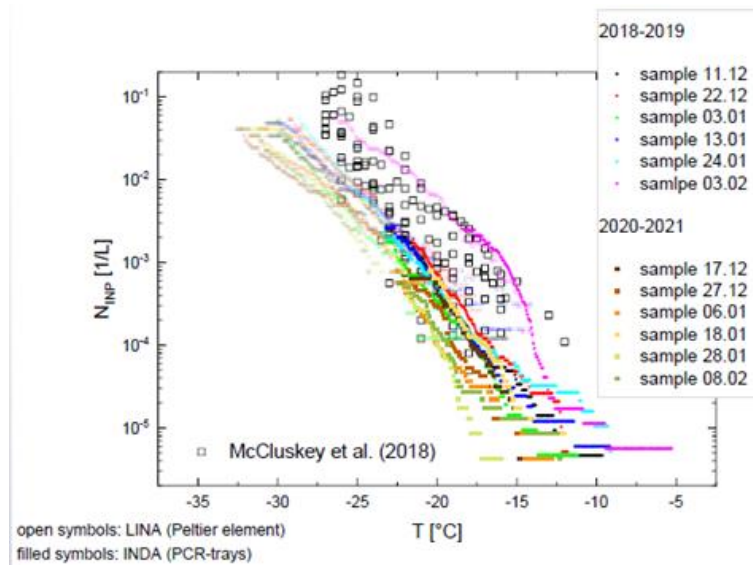


Figure 14: results of the laboratory analysis of INP filter samples taken during field seasons 2018/19 and 2020/21; analysed by TROPOS

This is in line with the findings of Tatzelt et al. (2022) who found that the Antarctic continent was found to be no important contributor to INP. They found similar INP values like McCluskey (2018) when the air masses had no terrestrial (i.e. non-Antarctic terrestrial) influence and higher INP concentrations when the air masses had terrestrial (i.e. other continents) influences. This information was used for the parameterisation of cloud and precipitation simulations, see respective sections in this report.

#### 4.2.3 Aerosol properties at PEA derived from remote sensing and from radiative transport modelling

In Figure 15 time series are shown of the AOD retrieved from the CIMEL measurements (Aeronet Aerosol Optical Depth v3 solar, level 1.5, <https://aeronet.gsfc.nasa.gov/> at 4 different wavelength: 340 nm, 380 nm, 440 nm and 500 nm, covering both the visible and the UV channel of the MAX-DOAS instrument) and AOD at 360 nm retrieved from the UV channel of the MAX-DOAS instrument with the default settings. Due to some instrumental issues in the beginning of the season 2020--2021, we also include MAX-DOAS retrievals with slightly different settings (mainly limited use of elevation angles) for that season. While we do not apply a thorough cloud filtering, we disregard profiles that have less than 60% of the total extinction in the lowermost 2 km.

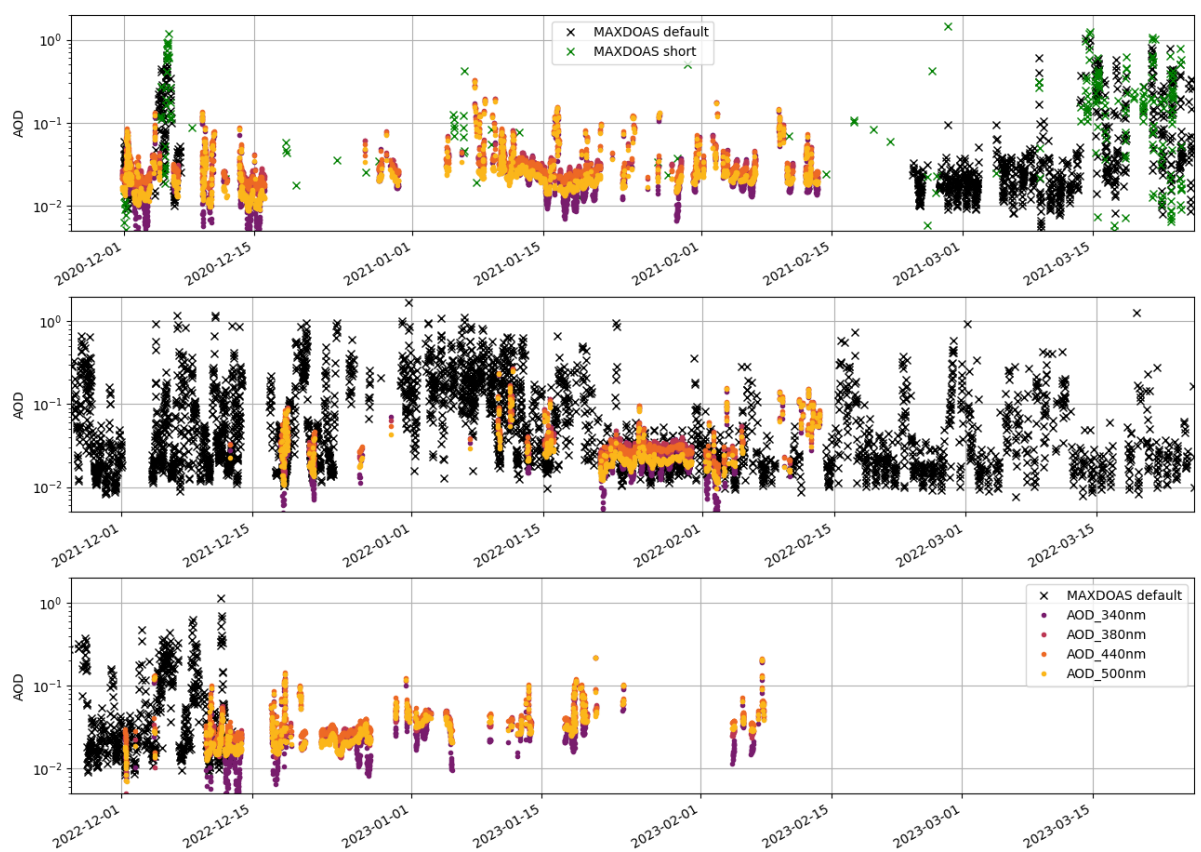


Figure 15: Time series of AOD from the Cimel instrument at 4 different wavelengths (see legend in the bottom panel) together with AODs retrieved from the MAX-DOAS UV channel. Note that due to instrumental problems in the first part of seasons 2020/2021, we also show AOD retrieval results with slightly different settings during that season. In terms of measurements, the scans used for the retrieval contain much fewer measurements and only include measurements at elevation angles above  $8^\circ$ , hence the scans are shorter.

As is apparent from Figure 15, there is quite some time overlap in data availability for the two instruments in season 2021/2022, but due to different instrumental issues as described in section 3.2.2, to a lesser degree during the other two seasons. Therefore, we concentrate the further analysis on that season and specifically on three sub-periods: 18 December 2021 - 21 December 2021, 10 January 2022 – 16 January 2022 and 21 January 2022 – 05 February 2022, as displayed in Figures 16 and 17, respectively in the UV and VIS channel.



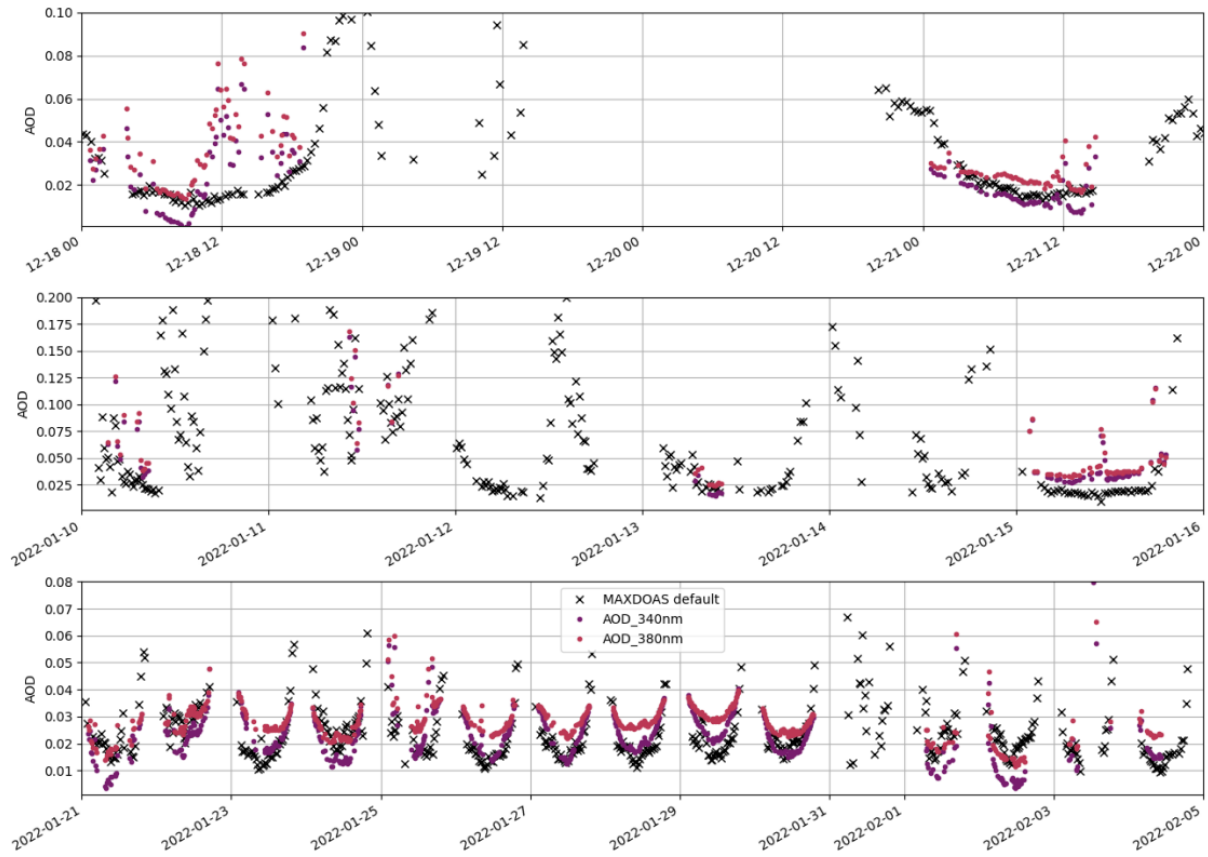


Figure 16: Time series of three periods in the season 2021/2022 during which both MAX-DOAS and CIMEL measurements are available and have valid measurements. Shown are the AOD from the CIMEL at wavelengths 340 nm and 380 nm, and the UV channel (360 nm) for the MAX-DOAS. Note the different y-scale for the different periods.

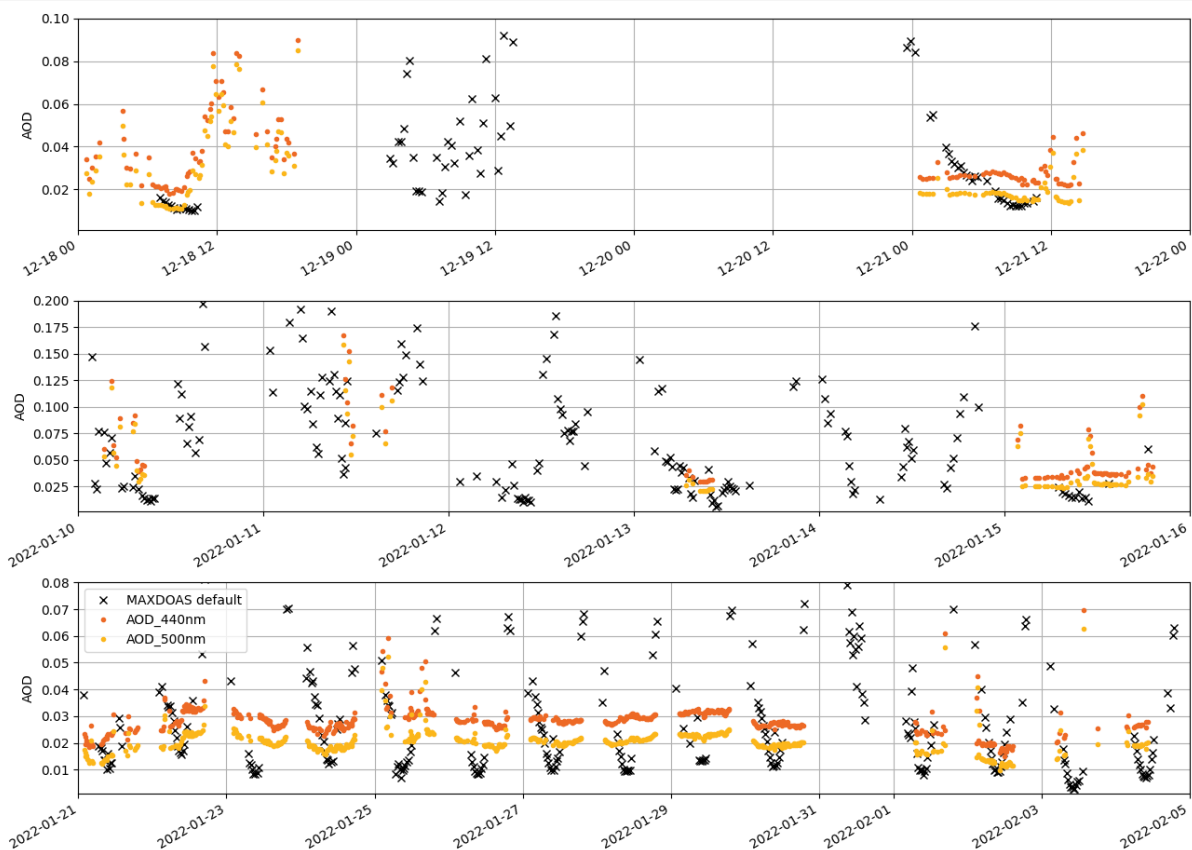


Figure.17: The same as Fig.2, but for CIMEL AOD at wavelengths 440 nm and 500 nm, and MAX-DOAS AOD retrieved from the visible wavelength window. (425 -- 490 nm).

In the UV (Fig. 16), the CIMEL and the MAXDOAS reproduced mostly the same diurnal patterns under clear sky conditions. It is further apparent that the AOD values retrieved from the UV window (338 -- 370 nm) often agreed better with the CIMEL 340 nm AODs, 340 nm being towards the short wavelength end of the used wavelength interval. On 18 December 2021 (upper panel), it might be the CIMEL detected some aerosol after 12 UT which were missed by the MAX-DOAS. It is important to note that the MAX-DOAS uses scattered light as source of information, while the CIMEL uses direct sunlight. The telescope azimuth angle of the telescope is fixed at  $17^\circ$  (i.e. approximately north north east) while the solar azimuth, and hence the direction of the Cimel between 12 and 17 UT is  $330^\circ$  --  $260^\circ$  (so north west by north to west by south). Therefore, the clear discrepancy in AOD values between roughly 12 and 17 UT does not necessarily point to a conceptual problem or a contradiction between the two measurements.

In the visible wavelength range, the diurnal variation, or the lack of it, was not well matched between the MAX-DOAS and the Cimel measurements (see Fig. 17). While during mid-day, the MAX-DOAS measurements severely underestimated the AOD compared to the Cimel values, the morning and evening MAX-DOAS retrieved values were too high. In general, the diurnal variation from the MAX-DOAS in the visible resembled more the variation seen in the UV for both the Cimel AODs and the MAX-DOAS AODs, while the Cimel AODs in the visible were very flat.

At the time of writing this report, investigations are ongoing to assess the quality of our AOD data, in particular of the CIMEL in the UV range and of the diurnal cycle of the MAX-DOAS AOD. The typical

AOD values of 0.02 to 0.04 are close to those previously reported in the literature in pristine Antarctica (Six et al., 2005, Tomasi et al., 2015).

#### 4.2.4 Implications and Recommendations

The MAX-DOAS measurements were discontinuous due to several issues, but they cover a full year of measurements, which make them interesting to study the seasonality of the AOD variations. To fully exploit the database, the cloud-filtering of the MAX-DOAS measurements must be improved. The cloud filtering is one subject of the FRM4DOAS activity (<https://frm4doas.aeronomie.be/>), which gathers international MAX-DOAS experts. Once finished, the work done on cloud filtering in FRM4DOAS should be applied to the CLIMB database. Beside the cloud filtering, the DOAS analysis and AOD retrievals could probably be further optimized also, to investigate e.g. the realism of the aforementioned diurnal variations.

The installed MAX-DOAS encountered several technical problems in the harsh conditions of Antarctica. Such an instrument is not easy to operate automatically in a station which is not inhabited and which may suffer power cuts. We recommend to replace it in the future by a zenith-only DOAS instruments, to focus on the stratosphere. Such an instrument would have fewer moving parts and its data quality would be less affected by ice on the windows. As noted in Section 4.6 below, the stratospheric dynamics in Antarctica in the very recent years motivates more measurements in this area.

The CIMEL operations went relatively smooth and the PEA crew is used to install and maintain it since 2009. This instrument could if possible come back at PEA every year without a dedicated scientists. The data quality in such a pristine environment should however be investigated with sunphotometer experts. For this the CLIMB database is interesting since we can compare the AOD from the CIMEL with those of the MAX-DOAS.

The found low concentrations of ice nucleating particles are an important finding for input data for the regional climate models for Antarctica. That the Antarctic continent itself seems to be no important contributor to INP is also relevant for further studies. In addition, the measurements of the optical particle sizer indicate that the concentration of particles larger than 500 nm (sizes relevant for INP) is very low in the interior of the Antarctic continent. It would be interesting to do more studies covering the whole year.

### 4.3 Characterisation of (semi-)volatile organic compounds

#### 4.3.1 Results of the 2019-2020 field campaign

The purpose-built autosampler proved capable of taking multiple samples in an extreme environment such as Antarctica during its deployment from December 2019 to October 2020. This led to, for the first time, a year-round dataset of 66 (O)VOCs reported for East-Antarctica and marks the first time that these compounds were successfully sampled in an automated sequential manner in Antarctica for later, off-line analysis. An in-situ breakthrough experiment was conducted to determine which compounds are sensitive to breakthrough. Samples were analyzed using the more traditional TD-GC-

MS and a novel technique encompassing a high-resolution PTR-Qi-TOFMS which was interfaced to a thermal desorber (TD-PTR-Qi-TOFMS). Characteristics of both instruments were compared and both techniques are found to be very complementary. While a good correlation was found between the concentrations of most compounds reported with both instruments, each proved to have its own advantages and drawbacks. The chromatographic separation of a GC proves to be a necessity to separate isomers (e.g., acetophenone and phenylacetaldehyde, being atmospheric oxidation products of ethylbenzene) and an EI-MS can detect alkanes and halocarbons. On the other hand, the alternative soft ionization source of the TD-PTR-Qi-TOFMS enabled to extend the range of measured analytes and the detection of some specific compounds such as DMS and 10 OVOCs related to atmospheric chemical processes which are rarely reported. Although the online capability of the PTR-Qi-TOFMS instrument was sacrificed to interface it with a thermal desorber for increased sensitivity, it still performs a fourfold faster compared to GC-MS, and the high-resolution MS adds an extra possibility for the screening of non-target compounds. The combination of both techniques and the non-target approach led to a very diverse dataset, spanning across different groups of organic compounds (aromatic, halocarbons,...), from very reactive to very persistent, and molecular weights from 46 to 253 g/mol. Concentration ranges from 0.5 ng/m<sup>3</sup> (methylbenzoate) to 20 µg/m<sup>3</sup> (benzoic acid) were found. Almost half (n=32) of the detected species in the samples were oxygenated compounds. Some (furaldehyde, furan, bromoform) show clear trends as a function of the amount of daylight (Figure 18). From literature, OVOCs could be identified as products from different atmospheric oxidation processes. Furthermore, several parent compounds (e.g. isoprene, toluene), as well as very advanced oxidation products (glyoxal), were found.

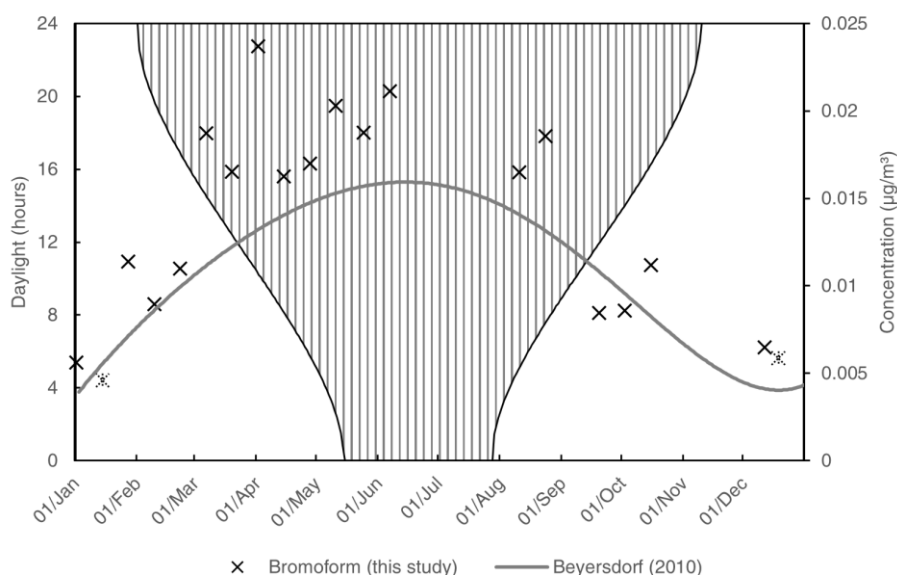


Figure 18: Observed trend of bromoform during the 2019-2020 sampling campaign

Through the development of a novel and robust sequential autosampler, and by using both instrumental analytical methods, year-round concentrations of 66 VOCs and atmospheric oxidation products were established, providing insight into the atmospheric chemistry in Antarctica.

### 4.3.2 Results from the 2020-2021 and 2021-2022 field campaign

During the 2020-2021 sample campaign, 2 autosamplers were deployed. The one installed at the station during the 2019-2020 was kept while a new one was installed at the CLIMB remote site. Both were charged with 25 samples and 5 blanks.

The CLIMB remote site posed a series of challenges. The instrument relied on a lead acid battery pack, which was charged with photovoltaic panels and a small wind turbine, built for this purpose. No remote communication was maintained with the instruments as there was no line-of-sight connection with the station. Finally, the weather conditions were even more extreme compared to the relatively mild microclimate around PEA. High wind speeds and extreme cold (down to  $-40^{\circ}\text{C}$ ) challenge both electronics as well as mechanical parts of the autosampler.

During the 2021-2022 the samples of the 2 autosamplers were collected. The station faced a black-out for a significant fraction of 2021 so the autosampler at the station only partially finished its programmed run. The autosampler at the CLIMB site got frozen shut after a few weeks of sampling only partially completing 2 samples. Furthermore, one of the charge controllers of the battery failed but battery power was maintained. During this campaign an upgraded version was installed with 50 samples at the CLIMB remote site. The power supply system got maintained and the faulty charge controller was replaced. The samples and the autosampler were collected by the IPF team during the 2022-2023 expedition.

### 4.3.3 Implications and Recommendations

With this unique dataset, we contribute to a scientific domain where in-situ collected data is very sparse. The developed technique offers potential to further investigate the atmospheric chemistry of organic compounds in remote areas. Especially a higher time resolution (50 sample positions) combined with the measurement of boundary conditions such as solar flux and particle number would lead to in-situ observations to further refine the current knowledge.

The measurement at the remote site proved a real challenge. While valuable knowledge was obtained about how to operate these types of instruments in these conditions, the failure rate remained quite high. It is recommended to use alternative battery chemistry, compatible to work in the extreme cold and to find reliable charging solutions. The used instruments should be further optimized to work autonomously without interaction of a user.

## 4.4 Meteorological analysis and dispersion modelling

### 4.4.1 Results from disdrometer, meteorological data loggers, radio soundings and AWS

#### 4.4.1.1 Results for the disdrometer

The disdrometer (Lufft WS100; Germany) for detecting precipitating and type (snow or drops) could finally be installed end of December 2021 on the southern roof of PEA, near to the micro-rain radar and ceilometer (Fig. 9). Unlike the micro-rain radar, it does not give vertical information. It detects only precipitation that reaches the disdrometer. All precipitation detected by the disdrometer was

classified as snow particles by the instrument. In Figure 19 some results from the disdrometer are shown. On the left graph of Figure 19 the cumulative precipitation (per day) over two months is illustrated as an example. It is clear that the precipitation occurred during some distinct events with long periods with no precipitation in between. On the right graph of Figure 19 the cumulative precipitation for one of these days (17-03-2022) is shown, illustrating the start of precipitation, the phase of strong precipitation and end of the event. These data will be compared to results of the micro-rain radar in more detail.

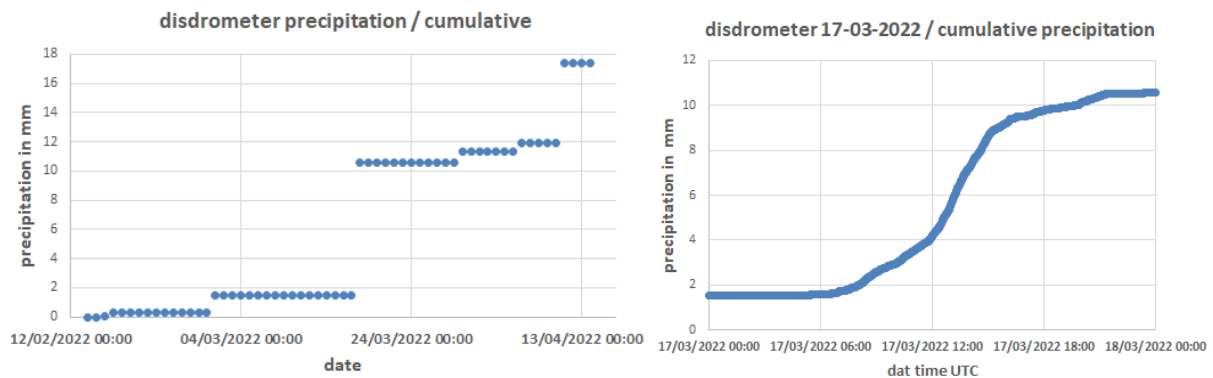
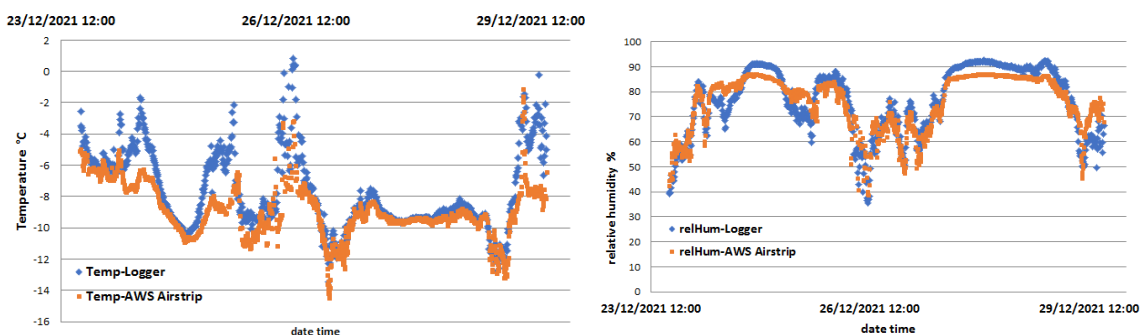


Figure 19: results from the disdrometer installed at PEA; left: cumulative precipitation (in mm) over a two month period; right: cumulative precipitation (in mm) for one day (17-03-2022)

#### 4.4.1.2 Results from the small meteorological data loggers

The robust small meteorological data loggers have been installed (i) on the roof of the southern science shelter; (ii) one on the Utsteinen nunatak, and (iii) at the Climb remote site (see Figs. 1 and 2). The logger at the remote site collected data until 25 April 2022, the one on the nunatak from 5 January 2022 to 5 January 2023, and the logger on the southern science shelter until 7 June 2022. In Figure 20, a one-week comparison of the logger from the science shelter with data from the AWS at the air strip is shown. Temperature showed clear differences, in particular for higher temperature peaks. Also the relative humidity values show differences. The pressure showed a distinct bias. It has to be noted that the data from the meteorological loggers are not yet corrected for the comparison with the standard sensors in the RMI laboratory. In addition, there might be a local meteorological effect due to the Utsteinen ridge, which might influence the wind field and flow and therefore also exert an influence on temperature and humidity. The pressure difference could be explained by the difference in altitude (around 70 m).



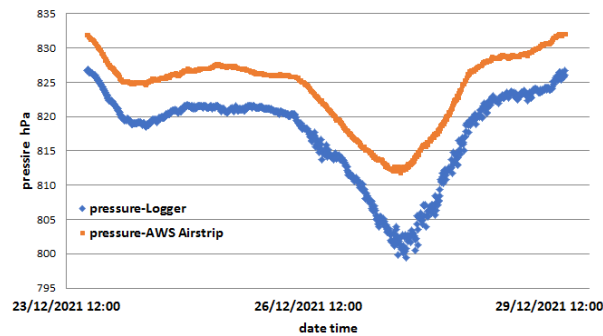


Figure 20: comparison of temperature, relative humidity and atmospheric pressure, measured with the small meteorological loggers (in blue) and data from the AWS at the air strip (around 2 km to the West).

From the three meteorological loggers, the temperature time series are shown in Figure 21. Not surprisingly, the remote site showed the lowest temperatures, no positive ones and with minima of  $-39^{\circ}\text{C}$ . The loggers at the sciences shelter and on the nunatak show some positive temperatures in the summer months and even in February. It is interesting to compare the data between the loggers from the science shelter and the nunatak. As the nunatak is 200 m higher than the Utsteinen ridge, the temperature on the nunatak can be expected to be lower. The comparison between the two loggers can give information how often there were temperature inversions.

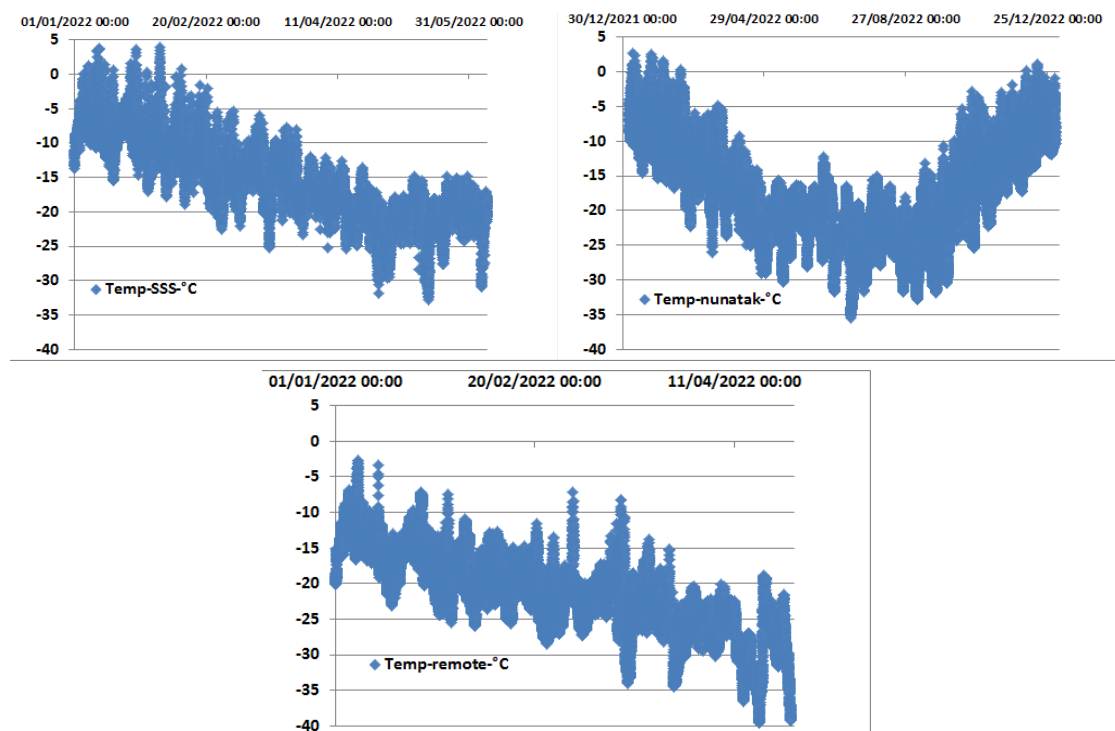


Figure 21: time series of temperature, measured with the meteorological data loggers, at three sites: southern science shelter (top left), nunatak summit (top right) and at the CLIMB remote site (bottom)

In Figure 22 it can be seen that this was indeed the case. Well into April, there are larger temperature differences between Utsteinen ridge and nunatak summit which cannot be explained by sensor differences or altitude difference only. There were several cases when temperature on the nunatak was more than  $4^{\circ}\text{C}$  warmer than on the ridge, and vice versa (with peaks of around  $8^{\circ}\text{C}$  colder and

10 °C warmer on the nunatak compared to the ridge). From April onwards, the temperature difference showed less variability, probably due to the less perturbed winter conditions (stronger circumpolar vortex, less synoptic weather systems reaching the Antarctic interior).

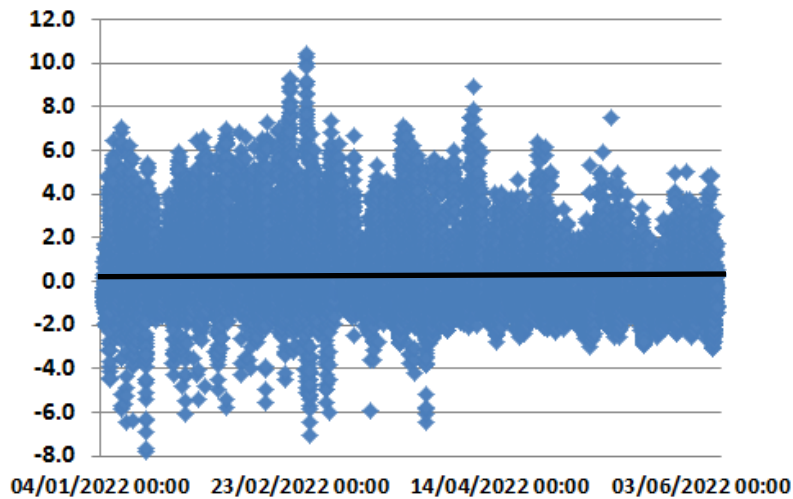


Figure 22: difference between the measured temperature at the southern science shelter and the nunatak summit (nunatak minus shelter; i.e. negative=colder at nunatak; positive=warmer at nunatak)

#### 4.4.1.3 Results from radio soundings and AWS

The radio sounding data has been analysed with respect to tropopause altitude, inversion layers, cloud layers, statistics of temperature, relative humidity, wind, and integrated water vapour. The tropopause altitude for the seasons up to 2021/22 is illustrated in Figure 23. The mean tropopause altitude at PEA was around 9000 m asl with some variability between the years. The years with longer than usual ozone holes (2020/21 and 2021/22) stand out not with a distinct difference in tropopause altitude, but with distinctly lower minimum temperatures at the tropopause. From the statistical analysis of the mean profiles, the altitude between 3000 to 4000 m asl stood out with respect to a peak in relative humidity, wind speed, the preferred occurrence of a cloud layer and of an inversion layer.

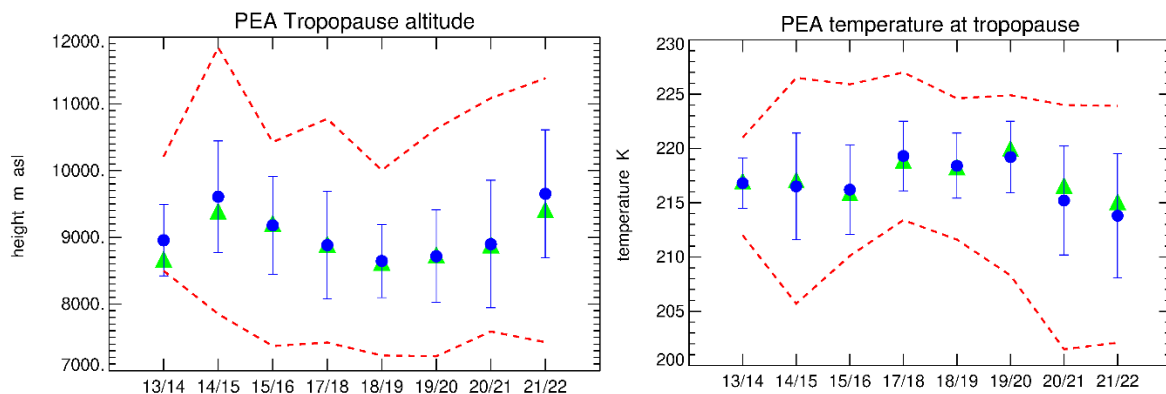


Figure 23: Tropopause at PEA measured by radio soundings; left: altitude above sea level; right: temperature at tropopause; blue: mean with standard deviation, green: median; red: minimum and maximum values



Further, from the humidity measurements of the soundings, the integrated water vapour (IWV) over the atmospheric column can be calculated. Values for the troposphere and the stratosphere are illustrated in Figure 24. The values for the troposphere compare well with similar studies for Antarctica. The values for the stratosphere should however be taken as indicative. The humidity sensor of the radiosonde is at its detection limit in the stratosphere and there is always the risk of icing of the sensor (in particular around the cold tropopause). After de-icing there might still be a hysteresis effect and therefore a wet bias. Although suspicious radio sounding profiles have been skipped, the high spread for minimum and maximum values indicate that the derived IWV values for the stratosphere are indicative-only.

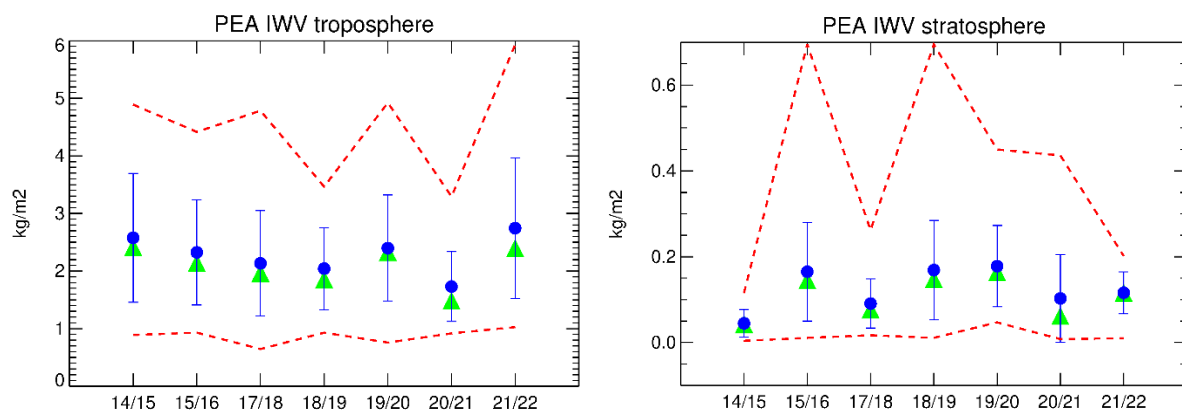


Figure 24: Integrated water vapour (IWV) measured at PEA by radio soundings; left: IWV within the troposphere; right: IWV within the stratosphere; blue: mean with standard deviation, green: median; red: minimum and maximum values

The AWS data has been processed by IMAU and is available via partners RMI or KUL. Alternative, current AWS data is available from the AWS near the Utsteinen air strip (IPF) and from the AWS east of Utsteinen ridge (EPFL, Lausanne, Switzerland). Data from the AWS near the air strip is currently available up to mid-February 2022.

#### 4.4.2 Source regions and transport pathways

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 25 below shows the four air mass origin clusters for the austral summer season (December-January-February) and autumn season (March-April-May). Similar, in Figure 26, the four air mass clusters are shown for austral winter (June-July-August) and spring (September-October-November).

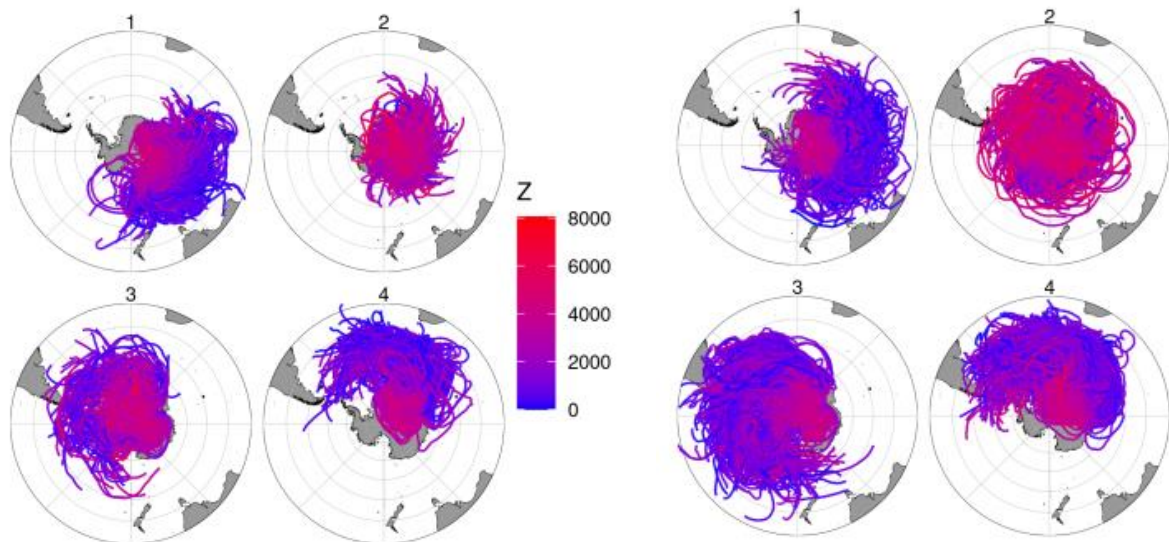


Figure 25: clusters of air mass origin, for austral summer (left) and austral autumn (right), for 11 years of backward trajectories (2010-2020) starting at PEA station; Z is height in m asl. All 10 days of the back trajectory calculation have been used for the clustering and all 10 days are included 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 the cluster 2, the source region was mostly restricted to the region above the Antarctic continent. The average altitude along the trajectories in this cluster was higher compared to the average altitude of air coming from source regions over the Southern Ocean, indicating that this cluster corresponded to air subsiding from aloft.

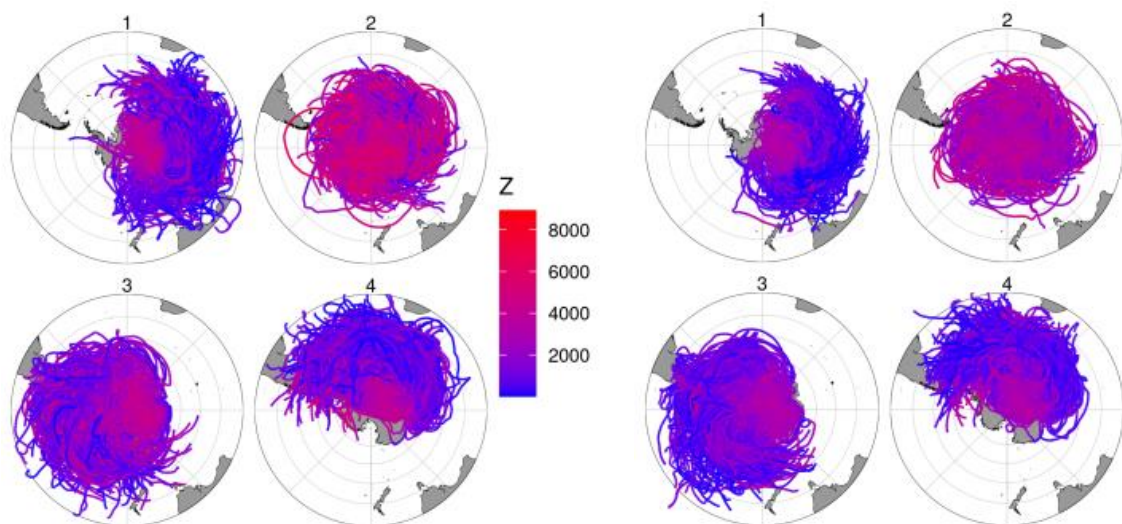


Figure 26: clusters of air mass origin, for austral winter (left) and austral spring (right), for 11 years of backward trajectories (2010-2020) starting at PEA station; Z is height in m asl. All 10 days of the back trajectory calculation have been used for the clustering and all 10 days are included in the graphs.

It is interesting to look at to what extent each cluster contributed to the total of backward trajectories per season. The percentages are given in Table 5. 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. During austral winter, the origins were more equally distributed. Between clusters 1, 3 and 4, the repartition was equal for all seasons, except for austral summer, when cluster 4 was under-represented.

Table 5: 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 27 shows the distribution of back trajectories, for the four seasons and between terrestrial or maritime origin. Terrestrial here means only Antarctica, no other continents. It clearly can be seen that the region of PEA was mostly influenced by air masses from Antarctica itself. This was most prominent during austral summer with more than 80% share of continental origin. Air masses with maritime origin had the highest share during austral winter. This is interesting because one would think that during austral summer when the circumpolar circulation is weaker there would be more influence from lower latitudes, thus also maritime origins; and vice versa for austral winter. It illustrates the importance of the sources of the Antarctic continent itself, related also to the subsidence of cold air masses also during austral summer, and the importance of synoptic weather patterns like cyclones during austral winter.

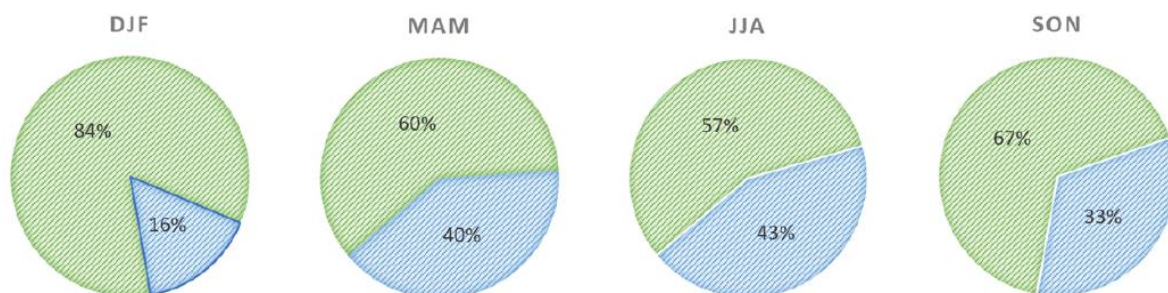


Figure 27: repartition of back trajectories, for the whole period 01/01/2010 to 31/12/2020, for the four seasons, and between continental (Antarctica; green) and maritime origin (blue)

Figure 28 shows the distribution of back trajectories between the four seasons, for distinct altitude sections, and for the four clusters. It can clearly be seen that again cluster 2 was dominating during all seasons and for the altitude sections above 1400 m asl. This in fact marks PEA station height and the lower altitude levels marked back trajectory origins from the coast upwards to the station.

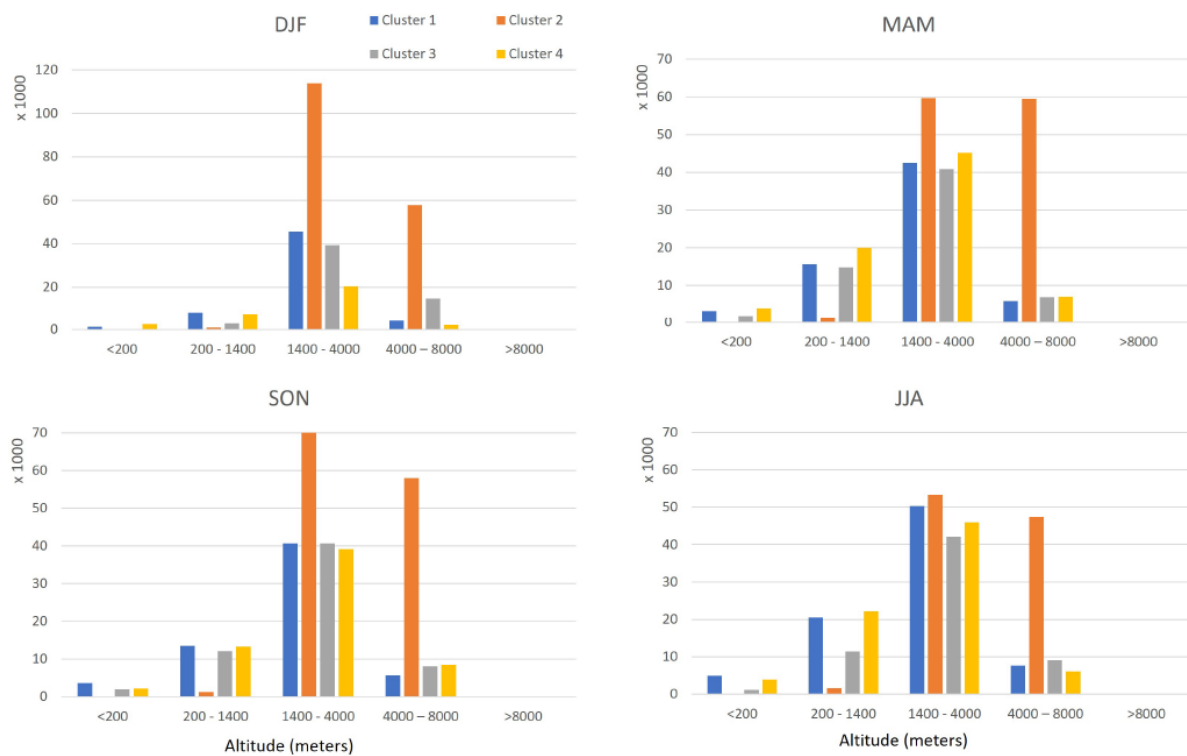


Figure 28: repartition of back trajectories, for the whole period 01/01/2010 to 31/12/2020, for the four seasons, the four clusters and for distinct altitude sections (altitude in m asl)

Figure 28 further shows that the altitude level above PEA up to 4000 m was dominating. The altitude levels with origins from the ocean and coast were of minor importance, as was the altitude level 4000 m to 8000 m. Higher levels were negligible.

#### 4.4.3 Implications and Recommendations

The disdrometer could detect the precipitation events. A comparison with the results from the micro-rain radar will be made in a follow-up project. The analysis of the radio soundings can rely now on already 8 summer seasons. Some characteristics with respect to tropopause altitude, integrated water vapour and distinct features like e.g., inversion layers or cloud layers start to have statistical value. As these soundings are in addition useful for weather forecasting and atmospheric re-analyses, they should be continued. A high variation of the temperature difference between the Utsteinen nunatak and ridge (around 200 m altitude difference) was found, higher than expected by the mere difference in height. This feature of local meteorology is very interesting and needs further investigation.

The results of the back trajectory and dispersion 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. Over all seasons, the Antarctic continent with air masses between 1400 m and 8000 m were dominating. Interestingly, the altitude level between 1400 and 4000 m was dominant. In the analysis of the radio soundings, the altitude level around 4000 m stood

out with respect to a peak in relative humidity, wind speed, the preferred occurrence of a cloud layer and of an inversion layer. It has to be noted that current atmospheric transport models have large uncertainties when simulating several weeks (> 4 weeks) of atmospheric transport what seems however to be necessary for transport from lower latitudes into Antarctica. Therefore, more elucidated modelling and research is needed how the East Antarctic atmosphere is connected to lower latitudes.

## 4.5 Regional Climate Modelling

### 4.5.1 Improvement of the surface mass balance and albedo in COSMO-CLM<sup>2</sup>

During the first project year, we contributed to an intercomparison study of five regional climate models for Antarctica (COSMO-CLM2, HIRHAM5, MAR3.10, MetUM and RACMO2.3p2; Mottram et al., 2021). The ensemble average of the surface mass balance of these five models is  $2329 \pm 94$  Gigatonnes (Gt) year<sup>-1</sup> over the common 1987–2015 period, with individual model results varying from  $1961 \pm 70$  to  $2519 \pm 118$  Gt year<sup>-1</sup>. Most of these differences occur in West Antarctica, the Peninsula and the transantarctic mountains. Such large differences are caused by different ice masks and different model resolution. Additionally, large differences for the integrated surface mass balance over the continent could be attributed to large differences in model precipitation at relatively few grid points in coastal areas. No systematic consistent trend was found in the surface mass balance over the ERA-Interim period.

### 4.5.2 Assessing the effect of aerosols on clouds and the climate of East Antarctica

In the setup tests for finding the optimal model settings to use, we first ran the model using a smaller grid (192 x 175 grid points) and forced it using the output from a coarser run. While the model was capturing the relevant microphysical effects as described later in this section, it was lacking in its representation of the cloud structure and occurrence times, compared to the ceilometer. An integration using the same grid, but forced by ERA-5 data directly, did not yield good results either, as the grid point containing the station, which we examined, was too close to the domain boundary. Thus, the domain size was increased to 392 x 392 grid points at the same 0.25° resolution, which fixed the issues with cloud structures, while maintaining a manageable computational load.

Using the modified version of COSMO-CLM<sup>2</sup> described earlier, we then created two case studies, where through varying the concentrations of INPs, we simulated both liquid- and non-liquid-containing clouds. The first study period is in the austral summer of 2011/12, spanning six weeks; the second one is in the austral winter of 2015, spanning three weeks. For these two case studies, we assessed the impact of the changed aerosol concentrations on cloud structure and phase (see Fig. 29). In both cases, we found the model to have an accurate representation of cloud height and occurrence, while the total vertical extent of the cloud could not be verified with the existing instrumentation. We found a strong anticorrelation between INP concentration and liquid water content, as was to be expected. This correlation also holds for the three INP concentrations representative for concentrations around PEA, however, the variability in cloud liquid water content within that range is not strong enough to have a significant impact on other climatic variables and the general cloud structure.

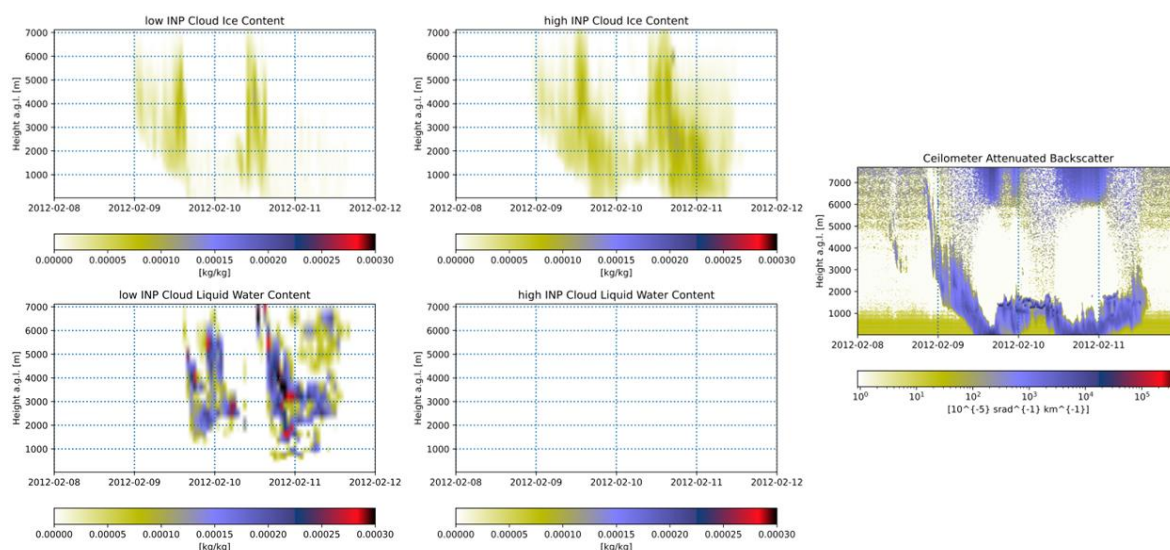


Figure 29: Vertical cross-sections of modelled and observed data at PEA over time. Top left: Cloud Ice Content, model output for  $5 \cdot 10^{-3}$  INP  $L^{-1}$ . Bottom left: Cloud Liquid Water Content, model output for  $5 \cdot 10^{-3}$  INP  $L^{-1}$ . Top middle: Cloud Ice Content, model output for  $2 \cdot 10^{-1}$  INP  $L^{-1}$ . Bottom middle: Cloud Liquid Water Content, model output for  $2 \cdot 10^{-1}$  INP  $L^{-1}$ . Right: Backscatter, as measured by the ceilometer. High backscatter values measured around the beginning of the 10<sup>th</sup> of February indicate the presence of liquid water.

The increase in liquid water in the cases with a lower INP concentration went together with a decrease in ice content. However, the increase in liquid water concentrations was stronger than the decrease in ice, thus a lower INP concentration meant a larger total cloud water content. As both liquid water and a higher total hydrometeor mass increased the reflectivity of clouds, a lower INP concentration caused an increase in the cloud radiative effect both for short- and longwave radiation. These opposing effects mostly cancelled out in the austral summer, while during the winter, the lack of incoming shortwave radiation meant a stronger warming effect through the increase in reflected longwave radiation.

#### 4.5.3 Implications and Recommendations

We were able to show that, as expected, the relative lack of INPs over Antarctica has significant implications for the phase of cloud water, which in turn has effects on the radiation balance, and that those changes can be simulated by the modified model. The computational complexity of the model limits its usability for long-term simulations however, especially if aerosols should be explicitly simulated. Thankfully though, our results indicate that the exact concentration of INPs, as long as it is within a realistic range for the region, is not strictly required for a good model performance. It would be interesting to see how a less computationally expensive version, which would have to be developed based on our obtained knowledge, would perform. A topic that has been left mostly unexplored is secondary ice formation, which is not fully implemented in our modified model, but which is known to have important effects on cloud structure and phase as well. Further investigation is also needed to differentiate the effects from the change in cloud phase on the radiative effects from the effects of the change in total water content.

## 4.6 Stratospheric Chemistry

Located between 10 and 50 km of altitude, the stratosphere is an essential component of our atmosphere since it hosts the ozone layer that protects life on earth from damaging UV radiations. In the eighties however, a phenomenon of ozone depletion was identified, which was attributed to the anthropogenic release of halogen-containing molecules (CFCs and related species) leading, once transported in the stratosphere, to catalytic ozone destruction. Although ozone depletion is a global problem in nature, particular conditions in polar regions lead an extreme phenomenon known as ozone hole. This effect is especially important in the Southern Hemisphere (Antarctica) because it is linked to the stratospheric temperature. Once temperatures drop below  $-78^{\circ}\text{C}$ , polar stratospheric clouds (PSCs) form, which exacerbate ozone depletion. In the Antarctic, the long presence of low temperatures is stimulating their formation, while in the Arctic larger year-to-year meteorological variability mitigates ozone depletion. The discovery of the ozone hole in 1985 (Farman et al., 1985) led to the rapid adoption of the Montreal Protocol in 1987 that bans the production of CFCs, halons, and other ozone-depleting chemicals. As a result, a stabilisation of the ozone levels could be reached in the nineties avoiding a major environmental catastrophe. However, because of the stability of the stratosphere against vertical motion, ozone-depleting substances only slowly escape from it and the recovery of the ozone hole to pre-1980 levels is only expected by around 2075 (Chipperfield et al., 2022). The monitoring of stratospheric ozone therefore remains very important especially in polar regions.

The chemical mechanisms that control the catalytic destruction of ozone in Antarctica are closely related to the formation of PSCs during polar night when temperatures drop below  $-78^{\circ}\text{C}$  (Solomon, 1999). Heterogeneous chemical reactions that convert reservoir molecules into reactive free radical molecules (such as ClO and Cl) then take place at the surface of the formed particles. Once the polar stratosphere is illuminated by the sun during austral Spring (late August – early September), these molecules are massively liberated and enter a series of catalytic reactions that destroy ozone very efficiently. Also the removal of  $\text{NO}_2$  molecules from ambient air due to their integration in PSCs particles, a process known as denoxification/denitrification, prevents the newly formed ClO to be converted back into its ClONO<sub>2</sub> reservoir, which maintains the ozone destruction mechanism active. In addition to their use to derive information on aerosol extinction (see section 4), MAX-DOAS measurements can also be exploited to infer the abundance of stratospheric ozone as well as a few key chemical species involved in the processes leading to the ozone hole. In particular, we focus here on the determination of ozone (Hendrick et al., 2011) and  $\text{NO}_2$  vertical columns (Hendrick et al., 2012) as well as OCIO slant columns (Pinardi et al., 2022). OCIO is formed by reaction of ClO and BrO radicals, it is therefore a direct indicator of the activation of chlorine species in the stratosphere. All these molecules are retrieved from zenith-sky spectra recorded at twilight when the observation geometry is such that the sensitivity of the measurements is strongly enhanced in the stratosphere. A side advantage of measuring at twilight is to allow for measurements at very low sun, so minimising the polar night gap.

Figure 30 displays time-series of daily measurements for the three target molecules covering a full annual cycle in 2022. Results illustrate well the chemical behaviour described above. One can see the OClO building up during the July-August period and, correspondingly, the ozone depletion starting in late August- early September and developing later to reach a minimum value of 126 DU on the 24<sup>th</sup> of October. The middle panel show the evolution of nitrogen dioxide ( $\text{NO}_2$ ) which, in the first part of the annual cycle, is mostly driven by the solar illumination which controls the photochemical equilibrium between  $\text{NO}_2$  and  $\text{NO}$ . From September onward, the impact of the PSC-induced denoxification/denitrification is clearly apparent. The denoxification persists until disappearance of the polar vortex, when polar air mixes with air from lower latitudes.

It is interesting to note that the ozone hole persisted until late November – early December. While the average ozone hole generally decreases quickly in October and even faster in November (see <https://atmosphere.copernicus.eu/three-peculiar-antarctic-ozone-hole-seasons-row-what-we-know>), the last three years have been characterised by an ozone hole persisting later in the season into November and closing well into December. Even though ozone depleting substances are on a decreasing trend since the late 1990s thanks to the Montreal Protocol, the occurrence of an extended ‘ozone hole season’ repeating itself 3 times in a row suggests that there could be changing factors in the South Pole stratosphere. The reasons behind this are not well understood, but they might be related to changes in the stratosphere dynamics.

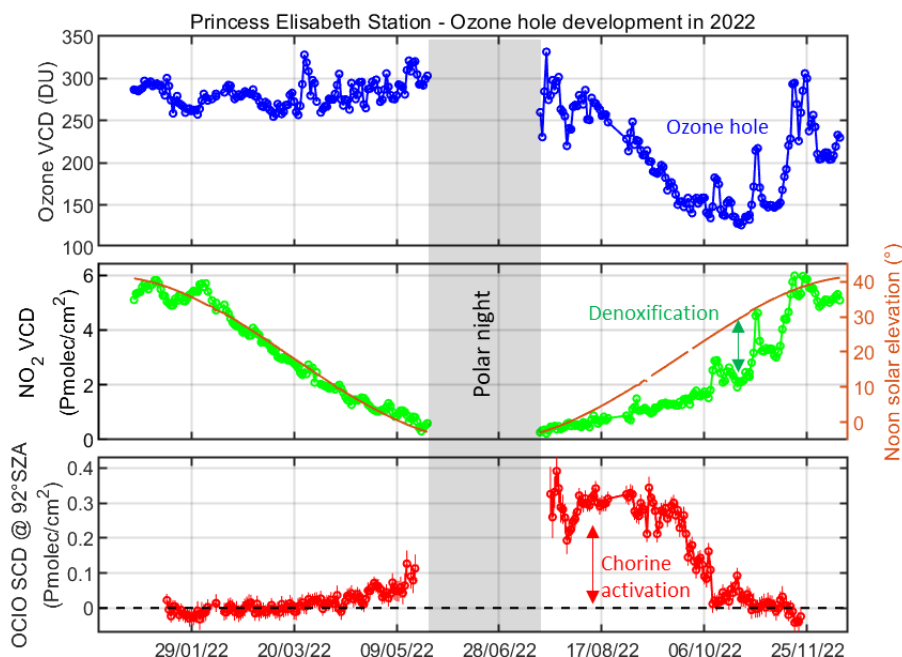


Figure.30: Time-series of ozone and  $\text{NO}_2$  vertical columns as well as OClO slant columns measured daily at twilight using the BIRA MAX-DOAS instrument installed at the PEA station in 2022. The data document well the annual evolution of stratospheric ozone together with the key chemical species involved in catalytic destruction processes active during the ozone hole period.



## 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 with all partners of the CLIMB project. Meetings were online during the Covid19-related period and in-person 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 held 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 are based on the collaboration between the different partners of CLIMB and have partners from the different institutes as co-author.

#### 5.1.2 Interaction with Follow-up Committee

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

There have been fruitful discussions between Nicole van Lipzig and several members of the follow-up committee (Prof. Michiel van den Broeke, Xavier Fettweis, Susanne Crewell) on regional climate modelling, remotes sensing of cloud and precipitation, and surface mass balance in polar regions. These discussions are continued. Alexander Mangold has been in contact with Heike Wex (Leibniz institute for tropospheric research, Leipzig, Germany) on the sampling of INP. Heike Wex is also involved in a scientific paper that is currently written by Florian Sauerland on the impact of ice nuclei concentrations on cloud properties and radiation in Antarctica. Prof. Dr. Frank Pattyn was part of the 2021-2022 Belare expedition when members of the CLIMB team were also present at the station. There have been extensive scientific discussions between Preben Van Overmeiren and Andy Delcloc with Frank Pattyn.

#### 5.1.3 Maintenance of observatory at Princess Elisabeth station

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

- In-person and virtual meetings and email-exchanges with the station operator and between CLIMB 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 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 (cloud and precipitation, aerosol in-situ, radio soundings)

Staff from the CLIMB partners was present at PEA and carried out the collection of samples and instrument operations during seasons 2020/2021 and 2021/2022. During season 2022/2023, no staff of CLIMB partners travelled to PEA. The following gives a brief description of the individual field expeditions:

- Season 2020/2021: 2 Pax present November-December 2020 and 1 Pax stayed until mid-January 2021 (Alexis Merlaud and Preben Van Overmeiren, respectively). They installed successfully CLIMB-related instrumentation at the station (MAXDOAS, CIMEL sunphotometer, aethalometer) and at the remote site (automated sampler for (S-)VOCs). They also helped to supervise and maintain the instrumentation for cloud and precipitation monitoring (ceilometer, micro-rain radar), aerosol monitoring (TEOM, nephelometer), helped with launching radio sondes. The micro-rain radar could be repaired and put back into operation with the help of the scientists at the station and the manufacturer. Together with the station operator, the final remote CLIMB site was explored and determined. A lot of work was done, with help of the technical team of the station, to successfully assemble the power supply for the instrumentation and to finally install and starting the automated (S-)VOC measurements on 2 January 2021 at that remote site.
- Season 2021/2022: 2 Pax present November-December 2021 (Preben Van Overmeiren and Andy Delcloo). They (re-)installed successfully CLIMB-related instrumentation at the station (CIMEL sunphotometer, MAXDOAS, disdrometer, meteo data logger, INP sampling) and at the remote site (automated sampler for VOCs, OPS, meteo data logger). This included also testing of the instruments and its set-up, and final installation at the remote site. They helped in addition to supervise and maintain the instrumentation for cloud and precipitation monitoring (ceilometer, micro-rain radar), aerosol monitoring (TEOM, aethalometer, nephelometer), ozone and uv monitoring (Brewer ozone spectrophotometer, pyranometers) and supported the station management with weather forecasts. The micro-rain radar dish was exchanged. At the end of season 2021/22, all instruments for whole-year operation were well running.
- Season 2022/2023 : NO Pax present, but station staff was trained to carry out the necessary tasks. The station team recovered the instrumentation and data from the CLIMB remote site (S-VOC sampler, OPS, meteo datalogger). Likewise, samples were recovered from the S-VOC sampler on the Northern Shelter and from the other meteo dataloggers. The MAXDOAS, CIMEL sunphotometer, ceilometer, micro-rain radar on the station roof were maintained as were the aerosol instruments in the specific southern shelter.

## 5.2 Data base and data dissemination

The ceilometer and MRR databases are maintained at KU Leuven and are available upon request. We contributed to a database of ceilometer measurements in Antarctica compiled by Bodeker Scientific, a research organization from New Zealand. Data of the radio soundings has been sent to the Global Telecommunications System (GTS) and is available via this channel. Data of the automatic weather station is available via partners KUL or RMI. The data of atmospheric aerosol physical properties will

be delivered to the EBAS data base. The CIMEL measurements as from 2009 are publicly available on the Aeronet website: [https://aeronet.gsfc.nasa.gov/cgi-bin/data\\_display\\_aod\\_v3?site=Utsteinen](https://aeronet.gsfc.nasa.gov/cgi-bin/data_display_aod_v3?site=Utsteinen). The MAX-DOAS measurements are available within the FRM4DOAS project team. In the context of this project, they will be made publicly available on NDACC data repository: <https://www-air.larc.nasa.gov/missions/ndacc/data.html>.

The COSMO-CLM<sup>2</sup> regional climate model full dataset is stored on a facility located at KU Leuven and is freely available for the scientific community and stakeholders on request. The full COSMO-CLM<sup>2</sup> output is too large in size to be put on an Open Access repository. Hence, access will be limited to requests by mail. Data for the atmospheric back trajectory and dispersion modelling is available via partner RMI. Likewise, for the COSMO model, it comprises a huge data amount, too large for Open Access repositories. The back trajectory and dispersion modelling data is stored on RMI servers.

The gathered chemical data within CLIMB 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.

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

Results of CLIMB have been widely disseminated and have been made public. In particular, see section 6.1 for peer-reviewed scientific publications, section 6.2 for presentations at scientific conferences, section 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). Nicole van Lipzig and 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, Christophe Walgraeve and Alexander Mangold 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. The follow-up committee member Tom Lachlan-Cope is chair of the SCAR Action Group ‘Antarctic Aerosols and Clouds’ which is promoting research and collaboration in the field of improved understanding of the aerosol-cloud interaction in Antarctica and in particular of improved parameterisations of clouds in regional climate models for Antarctica.

In addition to the contacts with the members of the Follow-up Committee, CLIMB partners have discussed the project and its outcomes with the scientific community as listed below:

- CLIMB partners participated in a polar symposium on 22 September 2022, organised by Belspo and the Royal Egmont institute for foreign affairs, BNCAR and APECS. Results from CLIMB have been presented there.
- Presentation of CLIMB results, Polar CORDEX meeting, September 2022, Bergen, Norway.
- CLIMB results were presented at the symposium on 'Belgian contributions to Earth Sciences in a Changing World' study day, organized by Belspo and the Belgian National Committee for Geodesy and Geophysics for the Belgian community in November 2022 in Brussels, Belgium.
- In 2022 Nicole van Lipzig received the prestigious Francqui Chair of the UC Louvain in recognition for her research and teaching activities. As part of this Chair, she held a lecture on Climate Modelling in Polar Regions at UC Louvain on 1st April 2022. In which she explained how climate models work and what we can learn from these models about climate change in polar region. CLIMB results were also included in this presentation. A recording can be found on: <https://youtu.be/kl4XvS-d4IE>
- Data of the radio soundings at PEA have been used in a paper on the physical habitat characterisation in the Antarctic Sør Rondane Mountains using satellite remote sensing by the Belgian Institute for Natural Sciences (in: Remote sensing Applications: Society and environment; <https://doi.org/10.1016/j.rsase.2021.100529>).
- Nicole van Lipzig contributed to news items for press, radio and television and she worked together with Frank Deboosere (of Flemish public broadcasting service) on a podcast on melting ice sheets, sea level rise and sea ice. She also gave lectures for the general public and non-scientific target groups. A list of these contributions is given in section 6.4
- In the process of developing the modified regional climate model, we were working closely together with Anna Possner (Goethe University, Frankfurt a.M., Germany), who wrote large parts of the code, and Kwinten van Weverberg (Ghent University, Belgium), who helped us with his expertise in aerosol chemistry and microphysics. Overall, the project helped tightening the link between the atmospheric modelling and atmospheric chemistry communities in Belgium.
- A seminar was held by Ruth Mottram from the Danish Meteorological Institute at KUL on the topic of 'Uncertainty and Challenges in Determining the Surface Mass Budget of Antarctica'.
- At KUL, there are currently two thesis students, with Maare Heene working on a bachelor thesis on *Linking atmospheric flow to cloud occurrence and properties in the vicinity of the Princess Elisabeth station, East Antarctica*; and Fleur De Wilder, who is preparing a master thesis titled *Linking Weather Observations at Princess Elisabeth Antarctica with Large-Scale Atmospheric Dynamics*. Both students analyse the measurement datasets from the cloud observatory in their theses.
- At RMI, one master student of University of Antwerp, Tom Stoops, did a 6-week internship in summer 2021 on the analysis of the radiosounding data at PEA. In summer 2022, a master student of Université Libre de Bruxelles, Charlotte Deramaix, analysed during an internship in detail the outcomes of the back trajectory simulations.

## 6. PUBLICATIONS

### 6.1 peer-reviewed scientific publications

- De Causmaecker, K., A. W. Delcloo, and A. Mangold (2022), Wildfire emissions and atmospheric dispersion, in: Mensink, C. Jorba, O. (eds), Air Pollution Modeling and its Application XXVIII. ITM 2021. Springer Proceedings in Complexity. Springer, Berlin, Heidelberg, <https://doi.org/10.1007/978-3-031-12786-1>
- 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](https://doi.org/10.1007/978-3-030-52171-4_18)
- Mottram, R., Hansen, N., Kittel, C., van Wessem, J. M., Agosta, C., Amory, C., Boberg, F., van de Berg, W. J., Fettweis, X., Gossart, A., van Lipzig, N. P. M., van Meijgaard, E., Orr, A., Phillips, T., Webster, S., Simonsen, S. B., and Souverijns, N.: What is the surface mass balance of Antarctica? An intercomparison of regional climate model estimates, *The Cryosphere*, 15, 3751–3784, <https://doi.org/10.5194/tc-15-3751-2021>, 2021
- Van Overmeiren P., Demeestere K., Mangold A., Delcloo A., Van Langenhove H., Walgraeve C., Year-round measurement of atmospheric volatile organic compounds using sequential sampling in Dronning Maud Land, East-Antarctica (2023) submitted to Atmospheric Environment (under review)
- Van Overmeiren P., Demeestere K., De Wispelaere P., Gili S., Mangold A., De Causmaecker, K., Mattielli, N., Delcloo A., Van Langenhove H., Walgraeve C., Four years of active sampling and measurement of atmospheric polycyclic aromatic hydrocarbons (PAHs) and oxygenated PAHs in Dronning Maud Land, East Antarctica (2023) submitted to Environmental Science and Technology (submitted).

### 6.2 Contributions to scientific conferences with proceedings

- Sauerland, F., Souverijns, N., Possner, A., Wex, H., Van Overmeiren, P., Mangold, A., Van Weverberg, K., and van Lipzig, N.: Simulating the effects of Ice-nucleating particles in Antarctica in COSMO-CLM<sup>2</sup>, EGU General Assembly 2023, Vienna, Austria, 24–28 Apr 2023, EGU23-14418, <https://doi.org/10.5194/egusphere-egu23-14418>, 2023.
- Sauerland, F., Souverijns, N., Mangold, A., Wex, H., Van Lipzig, N.: Cloud-Aerosol Interactions over Dronning Maud Land in COSMO-CLM<sup>2</sup>, Polar Cordex meeting 2022, Bergen, Norway, 28 to 30 Sep 2022.
- Mangold, A., K. De Causmaecker, Q. Laffineur, P. Van Overmeiren, C. Walgraeve, K. Demeestere, H. Van Langenhove, S. Boxho, N. Mattielli and A. Delcloo, Measurement of atmospheric particles and Volatile Organic Compounds at Princess Elisabeth Antarctica station: properties and air mass origin, 10<sup>th</sup> SCAR Open Science Conference 2022, 1-10 August 2022, virtual conference, India, 2022.
- Sauerland, F., Van Lipzig, N., Souverijns, N., Mangold, A., Van Overmeiren, P., and Wex, H.: Modelling the impact of cloud condensation and ice nuclei on the near-surface climate of Dronning Maud Land (East Antarctic) using the regional climate model COSMO-CLM<sup>2</sup>, EGU

- General Assembly 2022, Vienna, Austria, 23–27 May 2022, EGU22-12863, <https://doi.org/10.5194/egusphere-egu22-12863>, 2022.
- Van Overmeiren, P., Delcloo, A., De Causmaecker, K., Mangold, A., Demeestere, K., Van Langenhove, H., and Walgraeve, C.: Sequential sampling of Volatile Organic Compounds (VOCs) and atmospheric oxidation products in the Sør Rondane Mountains, East-Antarctica., EGU General Assembly 2022, Vienna, Austria, 23–27 May 2022, EGU22-12072, <https://doi.org/10.5194/egusphere-egu22-12072>, 2022.
  - Wex, H., Henning, S., Mangold, A., Van Overmeiren, P., Zeppenfeld, S., van Pinxteren, M., Herrmann, H., Dallosto, M., and Stratmann, F.: Ice Nucleating Particles in the Antarctic region, EGU General Assembly 2022, Vienna, Austria, 23–27 May 2022, EGU22-3796, <https://doi.org/10.5194/egusphere-egu22-3796>, 2022.
  - Merlaud, Alexis, Martina Friedrich, François Hendrick, Caroline Fayt, Christian Hermans, Alexander Mangold, Michel Van Roozendaal, MAX-DOAS O4 measurements under pristine aerosol-free conditions at the Belgian Princess Elisabeth Antarctic station, DOAS Workshop 2021, 11 May 2022, online.
  - 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, Bucharest, Romania, 20-24 September, 2021.
  - 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.
  - De Causmaecker, K., Delcloo, A., and A. Mangold: Identifying source regions for airborne particles in East Antarctica, Dronning Maud Land, using backward trajectory modelling, EGU General Assembly 2021, online, 19–30 Apr 2021, EGU21-14524, <https://doi.org/10.5194/egusphere-egu21-14524>, 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.

### 6.3 Outreach to the scientific community

- van Lipzig, N.P.M.: From Equator to Pole: Climate Change Processes and Impacts around the World, Inaugural lecture Francqui Chair Université catholique de Louvain, February 22, 2022, <https://youtu.be/GOD2RbJyODO>
- van Lipzig, N.P.M., 2022, Climate Modelling in Polar Regions. Topical lecture Francqui Chair Université catholique de Louvain March 18, 2022. <https://youtu.be/kl4XvS-d4IE>
- Van Lipzig, N. and A. Mangold, Climate research of Belgium in the Arctic and Antarctic, Polar symposium of Egmont Royal Institute for International Relations, 22 September 2022, Brussels, Belgium, 2022.

- Sauerland, F., Souverijns, N., Mangold, A., Wex, H., Van Lipzig, N.: Cloud-Aerosol Interactions in COSMO-CLM<sup>2</sup>, Study day on “Belgian contributions to Earth Sciences in a Changing World” organised by Belspo and the Belgian National Committee for Geodesy and Geophysics, Brussels, Belgium, 4 Nov 2022.
- Van Overmeiren, P., 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

- van Lipzig, N.P.M., 2023: Wetenschappelijke vooruitgang toont de noodzaak aan van snelle klimaatactie. Universiteit Derde Leeftijd Leuven vzw (UDLL), Leuven, 9 mei 2023
- 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
- van Lipzig, N.P.M., 2022. Scientific Advances Demonstrate Need for Rapid Climate Action. Public lecture Francqui Chair Université catholique de Louvain May 12, 2022. <https://youtu.be/hSRB7qScTmE>
- Van Lipzig, N.P.M., 2022: Klimaatverandering: een analyse. Lentecylus 2022:Koninklijke Vlaamse Academie van België (KVAB), Brussel, 04-05-2022. <https://youtu.be/nWBh7si9kok>
- Mangold, A., Science, Antarctica, Aerosol research – what it is about, Asgard X, European Space Education Resource Office, weather balloon launch competition for schools, talk and discussion with students from international secondary schools, 28 April 2022, Uccle, Belgium.
- van Lipzig, N.P.M., 2022. STEM - Kunnen we het klimaat van de toekomst voorspellen? Junior College Lectures - KU Leuven, 11/1/2022, <https://youtu.be/OgGzsgpefWA>
- van Lipzig, N.P.M., 2021. “Dissecting the COP26 - Was it successful?” Panel member in debate organized by the Green Office KU Leuven, 1 december 2021, Leuven, Belgium
- Presentation to the alumni organisation of the Ghent University bioscience engineering faculty (18-05-2021)
- Presentation on the Ghent University faculty research days by Preben Van Overmeiren (1/05/2021)
- Alexis Merlaud, BIRA-IASB Seminar, A trip to the ozone hole, 9 February 2021
- 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.
- van Lipzig, N.P.M., 2020. Klimaatverandering: De wetenschappelijke basis. Symposium on Climate change, organized by the Young Energy Reviewers Association, KU Leuven, 13/02/2020, Leuven, Belgium

### **Media presence (print, website, interviews)**

- Knack Interview: Klimatologe Nicole van Lipzig: ‘De spectaculaire daling van de kostprijs van zonne-energie is erg hoopgevend’ 24 October 2022 <https://www.knack.be/nieuws/milieu/klimaat/klimaatoptimisten/klimatologe-nicole-van-lipzig-de-spectaculaire-daling-van-de-kostprijs-van-zonne-energie-is-erg-hoopgevend/>
- Radio 1 “De Wereld Vandaag”: Nieuw klimaatrapport: “Corona heeft nauwelijks effect gehad in 2020 en 2021”, 10 Jan 2022, <https://radio1.be/luister/select/de-wereld-vandaag/nieuw-klimaatrapport-corona-heeft-nauwelijks-effect-gehad-in-2020-en-2021>
- Webnieuws (VRT): Laatste 7 jaar waren 7 warmste ooit gemeten op onze planeet, ongerustheid over methaan 10 Jan 2022, <https://www.vrt.be/vrtnws/nl/2022/01/10/laatste-7-jaar-waren-7-warmste-jaren-ooit-gemeten-wereldwijd/>
- Interviews with CLIMB partners on PEA station website or on website of International Polar Foundation : [http://www.antarcticstation.org/news\\_press/news\\_detail/andy\\_delcloo\\_and\\_preben\\_van\\_overmeiren\\_discuss\\_the\\_belspo\\_climb\\_project](http://www.antarcticstation.org/news_press/news_detail/andy_delcloo_and_preben_van_overmeiren_discuss_the_belspo_climb_project)
- [http://www.antarcticstation.org/news\\_press/news\\_detail/preben\\_van\\_overmeiren\\_explains\\_aerosols\\_and\\_cloud\\_formation\\_in\\_antarctica](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](http://www.antarcticstation.org/multimedia/picture_gallery/traverse_for_the_chase_and_climb_projects) ( 9 February 2021);
- En Antarctique, la station belge tourne en mode covid ; <https://dailyscience.be/23/12/2020/en-antarctique-la-station-belge-tourne-en-mode-covid/>
- A la station antarctique belge, la nouvelle saison scientifique démarre par une surveillance accrue de la couche d’ozone ; <https://dailyscience.brussels/fr/a-la-station-antarctique-belge-la-nouvelle-saison-scientifique-demarre-par-une-surveillance-accrue-de-la-couche-dozone/01/12/2020/>
- Expédition scientifique en Antarctique: une interview IASB ; <https://www.aeronomie.be/fr/nouvelles/2020/expedition-scientifique-en-antarctique-interview>
- Alexis Merlaud: Springtime ozone hole over Antarctica lasting longer this year; [http://www.antarcticstation.org/news\\_press/news\\_detail/alexis\\_merlaud\\_springtime\\_ozone\\_hole\\_over\\_antarctica\\_lasting\\_longer](http://www.antarcticstation.org/news_press/news_detail/alexis_merlaud_springtime_ozone_hole_over_antarctica_lasting_longer)
- [http://www.polarfoundation.org/news\\_press/news/alexander\\_mangold\\_contributions\\_of\\_research\\_to\\_polar\\_science\\_yopp\\_ipcc](http://www.polarfoundation.org/news_press/news/alexander_mangold_contributions_of_research_to_polar_science_yopp_ipcc) (22 July 2020)
- 2020: Nicole van Lipzig: Podcast – together with weatherman Frank Deboosere – melting ice sheets, sea level rise and sea ice. <https://www.vrt.be/vrtnws/nl/2020/10/15/planeet-frank-7/>

### **Videos/Movies/Podcasts**

- Interview met Pretty Smart Science (Isabel Torres) Podcast: modelling our changing climate, Nov 19, 2020. [https://www.youtube.com/watch?v=ymA7wuKHF\\_k](https://www.youtube.com/watch?v=ymA7wuKHF_k)
- van Lipzig, N.P.M., 2021. De relatie tussen extreem weer en klimaatverandering. Metaforum Middaggesprek, 2 december 2021, Leuven, Belgium,



[https://www.youtube.com/watch?app=desktop&v=KmmEp-11eic&list=PLb2yvGOMdiw\\_dKFDZYGrooPdaNE7DxGky&index=6](https://www.youtube.com/watch?app=desktop&v=KmmEp-11eic&list=PLb2yvGOMdiw_dKFDZYGrooPdaNE7DxGky&index=6)

### **Blogs:**

- RMI maintains a continuous blog on RMI's activities at Princess Elisabeth station: <https://belatmos.blogspot.com>
- 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**

- Alexis Merlaud, conférence Jeunesse et Sciences sur l'ozone, « Un trou dans un gaz », Louette Saint-Pierre, 25 Mars 2023
- KU Leuven Stories: Nicole Van Lipzig: "Elk tiende van een graad telt", 6 september 2022 <https://stories.kuleuven.be/nl/verhalen/nicole-van-lipzig-elk-tiende-van-een-graad-telt>
- 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
- Presentation of the scientific project to PEA crew by Andy Delcloo and Preben Van Overmeiren (12/01/2022)
- Alexis Merlaud: Atelier TADA a\_Schaerbeek sur la spectroscopie, l'ozone et l'Antarctique, 6 February 2021.
- 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.

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## 8. REFERENCES

- Agosta, C., Favier, V., Genthon, C., Gallée, H. Krinner, G., Lenaerts, J.T.M., van den Broeke, M.R. (2012). A 40-year accumulation dataset for Adelie Land, Antarctica and its application for model validation. *Climate Dynamics* 38, 75-86.
- Agosta, C. et al. (2019). Estimation of the Antarctic surface mass balance using the regional climate model MAR (1979-2015) and identification of dominant processes. *The Cryosphere* 13, 281-296.
- Ardon-Dryer, K., Z. Levin, and R. P. Lawson (2011), Characteristics of immersion freezing nuclei at the South Pole station in Antarctica, *Atmos. Chem. Phys.*, 11(8), 4015-4024, doi:10.5194/acp-11-4015-2011.
- Asmi, E. et al., Hygroscopicity and chemical composition of Antarctic sub-micrometre aerosol particles and observations of new particle formation, *Atmos. Chem. Phys.*, 10, 4253-4271, <https://doi.org/10.5194/acp-10-4253-2010>, 2010.
- Atkinson, R., Arey, J., 2003. Atmospheric Degradation of Volatile Organic Compounds. *Chem. Rev.* 103, 4605–4638. <https://doi.org/10.1021/cr0206420>
- Beirle, S., Dörner, S., Donner, S., Remmers, J., Wang, Y., and Wagner, T.: The Mainz profile algorithm (MAPA), *Atmos. Meas. Tech.*, 12, 1785–1806, <https://doi.org/10.5194/amt-12-1785-2019>, 2019.
- Bigg, E. K. (1973), Ice nucleus concentrations in remote areas, *J. Atmos. Sci.*, 30(6), 1153-1157, doi:10.1175/1520-0469(1973)030<1153:incira>2.0.co;2.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens, B., Zhang, X.Y., 2013. Clouds and Aerosols. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, in: Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V.B. and P.M.M. (Ed.), *Climate Change 2013 the Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 571–658. <https://doi.org/10.1017/CBO9781107415324.016>
- Bogumil, K., Orphal, J., and Burrows, J. P.: Temperature dependent absorption cross sections of O<sub>3</sub>, NO<sub>2</sub>, and other atmospheric trace gases measured with the SCIAMACHY spectrometer, *Proceedings of the ERS-Envisat-Symposium*, Goteborg, Sweden, 2000.
- Bromwich, D.H., Nicolas, J.P., Hines, K.M., Kay, J.E., Key, E.L., Lazzara, M.A., Libn, D., McFarquhar, G.M., Gorodetskaya, I.V., Grosvenor, D.P., Lachlan-Cope, T., van Lipzig, N.P.M. (2012). Tropospheric clouds in Antarctica. *Reviews of Geophysics* 50, RG1004.
- Chance, K. and R.L. Kurucz, An improved high-resolution solar reference spectrum for earth's atmosphere measurements in the ultraviolet, visible, and near infrared, *J. Quant. Spectroscopy Rad. Transfer*, 111(9), 1289-1295, <https://doi.org/10.1016/j.jqsrt.2010.01.036>, 2010.
- Chipperfield, M. P. and M. L. Santee (Lead Authors), S. Alexander, A. T. J. de Laat, D. G. Kinnison, J. Kuttippurath, U. Langematz, and K. Wargan. Polar Stratospheric Ozone: Past, Present, and Future, Chapter 4 in *Scientific Assessment of Ozone Depletion: 2022*, GAW Report No. 278, 509 pp., WMO, Geneva, 2022.
- Ciccioli, P., Cecinato, A., Brancaleoni, E., Frattoni, M., Bruner, F., Maione, M., 1996. Occurrence of Oxygenated Volatile Organic Compounds (VOC) in Antarctica. *Int. J. Environ. Anal. Chem.* 62, 245–253. <https://doi.org/10.1080/03067319608028137>

- Clarkson, T.S., Martin, R.J., 1997. Ethane and Propane in the southern marine troposphere 31, 3763–3771.
- Dall'Osto, M. et al., Antarctic sea ice region as a source of biogenic organic nitrogen in aerosols, *Sci. Rep.* 7, 6047, <https://doi.org/10.1038/s41598-017-06188-x>, 2017.
- Danckaert, T., Fayt, C., Van Roozendael, M., De Smedt, I., Letocart, V., Merlaud, A., and Pinaridi, G.: QDOAS Software user manual, Belgian Institute for Space Aeronomy, 2013
- DeFelice, T., V. Saxena and S. Yu, On the measurements of cloud condensation nuclei at Palmer station, Antarctica, *Atmos. Environ.*, 31, 4039–4044, 1997.
- Delcloo, A. W. and P. De Meutter (2019), Quantification of uncertainty in Lagrangian dispersion modelling, using ECMWF's new ERA5 ensemble, *Air Pollution Modeling and its Application XXVI*. DOI 10.1007/978-3-030-22055-6\_54.
- Delcloo, A.W., et al (2019). Spatio-Temporal Monitoring and Modelling of Birch Pollen in Belgium. *Air Pollution Modeling and its Application XXVI*. DOI 10.1007/978-3-030-22055-6\_12.
- De Meutter, P., Camps, J., Delcloo, A., and Termonia, P. (2018). Source localisation and its uncertainty quantification after the third DPRK nuclear test. *SCIENTIFIC REPORTS* 8. <http://dx.doi.org/10.1038/s41598-018-28403-z>
- DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., Niemand, M., Mohler, O., Snider, J. R., Wang, Z. et al.: Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles, *Atmospheric Chemistry and Physics*, 15, 393–409, 2015.
- Déry, S.J., Yau, M.K. (2002). Large-scale mass balance effects of blowing snow and surface sublimation. *Journal of Geophysical Research* 107, D23, 4679.
- Farman, J., Gardiner, B. and Shanklin, J. Large losses of total ozone in Antarctica reveal seasonal ClO<sub>x</sub>/NO<sub>x</sub> interaction. *Nature* 315, 207–210 (1985). <https://doi.org/10.1038/315207a0>
- Fiebig, M., D. Hirdman, C.R. Lunder, J.A. Ogren, S. Solberg, A. Stohl and R.L. Thompson, Annual cycle of Antarctic baseline aerosol: controlled by photooxidation-limited aerosol formation, *Atmos. Chem. Phys.*, 14, 3083–3093, <https://doi.org/10.5194/acp-14-3083-2014>, 2014.
- Fleischmann, O. C., M. Hartmann, J. P., Burrows, and J. Orphal, New ultraviolet absorption crosssections of BrO at atmospheric temperatures measured by time-windowing Fourier transform spectroscopy, *J. Photochem. Photobiol. A*, 168, 117–132, 2004.
- Forster, P., Storelvmo, T., Armour, K., Collins, W., Dufresne, J.-L., Frame, D., Lunt, D.J., Mauritsen, T., Palmer, M.D., Watanabe, W., Wild, M., Zhang, H., 2021. The Earth's Energy Budget, Climate Feedbacks, and Climate Sensitivity. In *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. <https://doi.org/10.1017/9781009157896.009>
- Friedrich, M. M., Rivera, C., Stremme, W., Ojeda, Z., Arellano, J., Bezanilla, A., García-Reynoso, J. A., and Grutter, M.: NO<sub>2</sub> vertical profiles and column densities from MAX-DOAS measurements in Mexico City, *Atmos. Meas. Tech.*, 12, 2545–2565, <https://doi.org/10.5194/amt-12-2545-2019>, 2019.
- Frieß, U., Beirle, S., Alvarado Bonilla, L., Bösch, T., Friedrich, M. M., Hendrick, F., Piders, A., Richter, A., van Roozendael, M., Rozanov, V. V., Spinei, E., Tirpitz, J.-L., Vlemmix, T., Wagner, T., and Wang, Y.: Intercomparison of MAX-DOAS vertical profile retrieval algorithms: studies using synthetic data, *Atmos. Meas. Tech.*, 12, 2155–2181, <https://doi.org/10.5194/amt-12-2155-2019>, 2019.

- Gallée, H., et al. (2015). Characterisation of the boundary layer at Dome C (East Antarctica) during the OPALE summer campaign. *Atmospheric Chemistry and Physics* 15, 6225-6236.
- Girard, E., Blanchet, J.P. (2001). Simulation of Arctic diamond dust, ice fog, and thin stratus using an explicit aerosol-cloud-radiation model. *Journal of the Atmospheric Sciences* 58, 1199-1221.
- Giorgi, F., Gutowski, W.J. (2015). Regional dynamical downscaling and the CORDEX initiative. *Annual Review of Environment and Resources* 40, 467-490.
- Gong, X., H. Wex, M. van Pinxteren, N. Triesch, K. W. Fomba, J. Lubitz, C. Stolle, B. Robinson, T. Müller, H. Herrmann, and F. Stratmann (2019), Characterization of aerosol particles at Cape Verde close to sea and cloud level heights - Part 2: ice nucleating particles in air, cloud and seawater, *Atmos. Chem. Phys. Discuss.*, submitted to, doi:10.5194/acp-2019-xxx.
- Gonzales-Toril, E.R. et al., Bacterial diversity of autotrophic enriched cultures from remote, glacial Antarctica, Alpine and Andean aerosol, snow and soil samples, *Biogeosciences*, 6, 33-44, 2009.
- Gorodetskaya, I.V., van Lipzig, N.P.M., van den Broeke, M.R., Mangold, A., Boot, W., Reijmer, C.H. (2013). Meteorological regimes and accumulation patterns at Utsteinen, Dronning Maud Land, East Antarctica: analysis of two contrasting years. *Journal of Geophysical Research: Atmospheres* 118, 1700-1715.
- Gorodetskaya, I.V., Tsukernik, M., Claes, K., Ralph, M.F., Neff, W.D., van Lipzig, N.P.M. (2014). The role of atmospheric rivers in anomalous snow accumulation in East Antarctica. *Geophysical Research Letters* 16, 6199-6206.
- Gorodetskaya I., Kneifel S., Maahn M., Van Tricht K., Thiery W., Schween J., Mangold A., Crewell S., Van Lipzig N. (2015). Cloud and precipitation properties from ground-based remote-sensing instruments in East Antarctica. *Cryosphere*, 9, 285-304. doi: 10.5194/tc-9-285-2015.
- Hamilton, D.S., L.A. Lee, K.J. Pringle, C.L. Reddington, D.V. Spracklen and K.S. Carslaw, Occurrence of pristine aerosol environment on a polluted planet, *Proc. Natl. Acad. Sci. USA*, 111, 18466-18471, <https://doi.org/10.1073/pnas.1415440111>, 2014.
- Gossart A., Souverijns N., Gorodetskaya IV., Lhermitte S., Lenaerts JT M., Schween JH., Mangold A., Laffineur Q., Van Lipzig N. (2017). Blowing snow detection from ground-based ceilometers: application to East Antarctica. *Cryosphere*, 11 (6), 2755-2772. doi: 10.5194/tc-11-2755-2017.
- Griffiths, P.T., Murray, L.T., Zeng, G., Shin, Y.M., Abraham, N.L., Archibald, A.T., Deushi, M., Emmons, L.K., Galbally, I.E., Hassler, B., Horowitz, L.W., Keeble, J., Liu, J., Moeini, O., Naik, V., O'Connor, F.M., Oshima, N., Tarasick, D., Tilmes, S., Turnock, S.T., Wild, O., Young, P.J., Zanis, P., 2021. Tropospheric ozone in CMIP6 simulations. *Atmos. Chem. Phys.* 21, 4187–4218. <https://doi.org/10.5194/acp-21-4187-2021>.
- Guyot, A., Protat, A., Alexander, S. P., Klekociuk, A. R., Kuma, P., and McDonald, A. (2022). Detection of supercooled liquid water containing clouds with ceilometers: development and evaluation of deterministic and data-driven retrievals. *Atmospheric Measurement Techniques*, 15(12), 3663–3681. <https://doi.org/10.5194/amt-15-3663-2022>.
- Hartmann, M., T. Blunier, S. O. Brügger, J. Schmale, M. Schwikowski, A. Vogel, H. Wex, and F. Stratmann (2019), Variation of ice nucleating particles in the European Arctic over the last centuries, *Geophys. Res. Lett.*, 46, *accepted*, doi:10.1029/2019GL082311.
- Hendrick, F., Mahieu, E., Bodeker, G. E., Boersma, K. F., Chipperfield, M. P., De Mazière, M., De Smedt, I., Demoulin, P., Fayt, C., Hermans, C., Kreher, K., Lejeune, B., Pinardi, G., Servais, C., Stübi, R., van der A, R., Vernier, J.-P., and Van Roozendaal, M.: Analysis of stratospheric NO<sub>2</sub> trends above Jungfraujoch using ground-based UV-visible, FTIR, and satellite nadir observations, *Atmos. Chem. Phys.*, 12, 8851–8864, <https://doi.org/10.5194/acp-12-8851-2012>, 2012.

- Hendrick, F., Pommereau, J.-P., Goutail, F., Evans, R. D., Ionov, D., Pazmiño, A., Kyrö, E., Held, G., Eriksen, P., Dorokhov, V., Gil, M., and Van Roozendaal, M.: NDACC/SAOZ UV-visible total ozone measurements: improved retrieval and comparison with correlative ground-based and satellite observations, *Atmos. Chem. Phys.*, 11, 5975–5995, <https://doi.org/10.5194/acp-11-5975-2011>, 2011.
- Herenz, P., H. Wex, A. Mangold, Q. Laffineur, I. V. Gorodetskaya, Z. L. Flemming, 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:10.5194/acp-19-275-2019.
- Hönninger, G., von Friedeburg, C., and Platt, U.: Multi axis differential optical absorption spectroscopy (MAX-DOAS), *Atmos. Chem. Phys.*, 4, 231–254, <https://doi.org/10.5194/acp-4-231-2004>, 2004.
- Kim, J., Y.I. Yoon, Y. Gim, H.J. Kang, J.H. Choi, K.-T. Park and B.Y. Lee, Seasonal variations in physical characteristics of aerosol particles at the King Sejong Station, Antarctic Peninsula, *Atmos. Chem. Phys.*, 17, 12985-12999, <https://doi.org/10.5194/acp-17-12985-2017>, 2017.
- Koponen, I.K., A. Virkkula, R. Hillamo, V.M. Kerminen and M. Kulmala, Number size distribution and concentrations of the continental summer aerosol in Queen Maud Land, Antarctica, *J. Geophys. Res.*, 108, 4587, <https://doi.org/10.1029/2003jd003614>, 2003.
- Kyrö, E. et al., Antarctic new particle formation from continental biogenic precursors, *Atmos. Chem. Phys.*, 13, 3527-3546, <https://doi.org/10.5194/acp-13-3527-2013>, 2013.
- Lachlan-Cope, T. (2010). Antarctic clouds. *Polar Research* 29, 150-158.
- Lachlan-Cope, T., Listowski, C., O’Shea, S. (2016). The microphysics of clouds over the Antarctic Peninsula – part 1: observations. *Atmospheric Chemistry and Physics* 16, 15605-15617.
- Langen, P.L., Fausto, R.S., Vandecrux, B., Mottram, R.H., and Box, J.E.: Liquid water flow and retention on the Greenland ice sheet in the regional climate model HIRHAM5: Local and large-scale impacts, *Front. Earth Sci.*, 4, 110, <https://doi.org/10.3389/feart.2016.00110>, 2017.
- Lenaerts, J.T.M. et al. (2018). Climate and surface mass balance of coastal West Antarctica resolved by regional climate modelling. *Annals of Glaciology* 59, 29-41.
- Levenberg, K.: A method for the solution of certain non-linear problems in least squares, *Quart. Appl. Math.* 2 (1944), 164-168, Primary 65.0X, <https://doi.org/10.1090/qam/10666>
- Ligtenberg, S.R.M., van de Berg, W.J., van den Broeke, M.R., Rai, J.G.L., van Meijgaard, E. (2013). Future surface mass balance of the Antarctic ice sheet and its influence on sea level change, simulated by a regional atmospheric climate model. *Climate Dynamic* 41, 867-884.
- Listowski, C., Lachlan-Cope, T. (2017). The microphysics of clouds over the Antarctic Peninsula – Part 2: modelling aspects within Polar WRF. *Atmospheric Chemistry and Physics* 17, 10195-10221.
- Lucas-Picher, P., Wulff-Nielsen, M., Christensen, J.H., Aðalgeirsdóttir, G., Mottram, R., and Simonsen, S.B.: Very high resolution regional climate model simulations over Greenland: Identifying added value, *J. Geophys. Res.-Atmos.*, 117, <https://doi.org/10.1029/2011JD016267>, 2012.
- Maahn, M., Burgard, C., Crewell, S., Gorodetskaya, I.V., Kneifel, S., Lhermitte, S., Van Tricht, K., van Lipzig, N.P.M. (2014). How does the spaceborne radar blind zone affect derived surface snowfall statistics in polar regions? *Journal of Geophysical Research: Atmospheres* 119, 13604-13620.
- Matus, A.V., L’Ecuyer, T.S. (2017). The role of cloud phase in Earth’s radiation budget. *Journal of Geophysical Research: Atmospheres* 122, 2559-2578.
- McCluskey, C. S., T. C. J. Hill, R. S. Humphries, A. M. Rauker, S. Moreau, P. G. Strutton, S. D. Chambers, A. G. Williams, I. McRobert, J. Ward, M. D. Keywood, J. Harnwell, W. Ponsonby, Z. M. Loh, P. B. Krummel, A. Protat, S. M. Kreidenweis, and P. J. DeMott (2018), Observations of Ice Nucleating

- Particles Over Southern Ocean Waters, *Geophys. Res. Lett.*, 45(21), 11989-11997, doi:10.1029/2018gl079981.
- Meller, R. and G. K. Moortgat, Temperature dependence of the absorption cross sections of formaldehyde between 223 and 323K in the wavelength range 225–375 nm, *J. Geophys. Res.*, 105, 7089–7101, 2000.
- Mottram, R., Hansen, N., Kittel, C., van Wessem, J. M., Agosta, C., Amory, C., Boberg, F., van de Berg, W. J., Fettweis, X., Gossart, A., van Lipzig, N. P. M., van Meijgaard, E., Orr, A., Phillips, T., Webster, S., Simonsen, S. B., and Souverijns, N.: What is the surface mass balance of Antarctica? An intercomparison of regional climate model estimates, *The Cryosphere*, 15, 3751–3784, <https://doi.org/10.5194/tc-15-3751-2021>, 2021.
- Newman, A.J., Kucera, P.A., Bliven, L.F., 2009. Presenting the Snowflake Video Imager (SVI). *Journal of Atmospheric and Oceanic Technology* 26, 167–179. doi:10.1175/2008JTECHA1148.1.
- O’Shea, S.J. et al., In situ measurements of cloud microphysics and aerosol over coastal Antarctica during the MAC campaign, *Atmos. Chem. Phys.*, 17, 13049-13070, <https://doi.org/acp-17-13049-2017>, 2017.
- O’Sullivan, D., M. P. Adams, M. D. Tarn, A. D. Harrison, J. Vergara-Temprado, G. C. E. Porter, M. A. Holden, A. Sanchez-Marroquin, F. Carotenuto, T. F. Whale, J. B. McQuaid, R. Walshaw, D. H. P. Hedges, I. T. Burke, Z. Cui, and B. J. Murray (2018), Contributions of biogenic material to the atmospheric ice-nucleating particle population in North Western Europe, *Scientific Reports*, 8, 13821, doi:10.1038/s41598-018-31981-7.
- Ovadnevaite, J., et al. (2017), Surface tension prevails over solute effect in organic-influenced cloud droplet activation, *Nature* (546), 637-641.
- Palerme, C., Kay, J.E., Genthon, C., L’Ecuyer, T.L., Wood, N.B., Claud, C. (2014). How much snow falls on the Antarctic ice sheet? *The Cryosphere* 8, 1577-1587.
- Pattyn, F., et al. (2018). The Greenland and Antarctic ice sheets under 1.5°C global warming. *Nature Climate Change* 8, 1053-1061.
- Philips, E., 2019. Snowfall over Antarctica: what are we measuring. Master thesis Geography KULeuven June 2019, N.P.M. van Lipzig (supervisor), N. Souverijns (co-supervisor), 85 pp.
- Pinardi, G., Van Roozendaal, M., Hendrick, F., Richter, A., Valks, P., Alwarda, R., Bogner, K., Frieß, U., Granville, J., Gu, M., Johnston, P., Prados-Roman, C., Querel, R., Strong, K., Wagner, T., Wittrock, F., and Yela Gonzalez, M.: Ground-based validation of the MetOp-A and MetOp-B GOME-2 OClO measurements, *Atmos. Meas. Tech.*, 15, 3439–3463, <https://doi.org/10.5194/amt-15-3439-2022>, 2022.
- Platt, U., Stutz, J.; *Differential Optical Absorption Spectroscopy, Physics of Earth and Space Environments*, Springer-Verlag Berlin, Heidelberg, <https://doi.org/10.1007/978-3-540-75776-4>, 2008.
- Pöhlker, M.L. et al., (2016), Long-term observations of cloud condensation nuclei in the Amazon rain forest – Part 1: Aerosol size distribution, hygroscopicity and new model parameterizations for CCN prediction, *Atmos. Chem. Phys.* 16, 15709-15740, doi.org/acp-16-15709-2016.
- Possner, A., Ekman, A. M., & Lohmann, U. (2017). Cloud response and feedback processes in stratiform mixed-phase clouds perturbed by ship exhaust. *Geophysical Research Letters*, 44(4), 1964-1972.
- Rodgers, C. D.: *Inverse methods for atmospheric sounding: theory and practice*, World Scientific Publishing, London, 2000.
- Rosenfeld, D., Lohmann, U., Raga, G.B., O’Dowd, C.D., Kulmala, M., Fuzzi, S., Reissell, A., Andreae, M.O. (2008). *Science* 321, 1309-1313.

- Rosenfeld, D., Sherwood, S., Wood, R., Donner, L. (2014). Climate effects of aerosol-cloud interactions. *Science* 343, 379-380.
- Rothman, L. S., I.E. Gordon, R.J. Barber, H. Dothe, R.R. Gamache, A. Goldman, V.I. Perevalov, S.A. Tashkun, and J. Tennyson, HITEMP, the high-temperature molecular spectroscopic database, *J. Quant. Spectrosc. Radiat. Transfer*, 111, 2139-2150, 2010.
- Rudolph, J., Khedim, A., Wagenbach, D., 1989. The seasonal variation of light nonmethane hydrocarbons in the Antarctic troposphere. *J. Geophys. Res. Atmos.* 94, 13039–13044. <https://doi.org/10.1029/JD094iD10p13039>
- Rudolph, J., Khedim, A., Clarkson, T., Wagenbach, D., 1992. Long-term measurements of light alkanes and acetylene in the Antarctic troposphere. *Tellus B Chem. Phys. Meteorol.* 44, 252–261. <https://doi.org/10.3402/tellusb.v44i4.15453>
- Sanchez, K., G.C. Roberts, M.D. Keywood and R. Humphries (2018), Measured and simulated cloud condensation nuclei over the Southern Ocean and their effects on cloud microphysical and optical properties, AGU Fall Meeting, 2018, abstract #A11B-02.
- Schmale, J., J. Schneider, E. Nemitz, Y.S. Tang, U. Dragosits, T.D. Blackall, P.N. Trathan, G.J. Phillips, M. Sutton and C.F. Braban, Sub-Antarctic marine aerosol: dominant contributions from biogenic sources, *Atmos. Chem. Phys.*, 13, 8669-8694, <https://doi.org/acp-13-8669-2013>, 2013.
- Seifert, A., Beheng, K.D. (2006). A two-moment cloud microphysics parameterization for mixed-phase clouds. Part 1: model description. *Meteorology and Atmospheric Physics* 92, 45-66.
- Serdyuchenko, A., Gorshchev, V., Weber, M., Chehade, W., and Burrows, J. P.: High spectral resolution ozone absorption cross-sections – Part 2: Temperature dependence, *Atmos. Meas. Tech.*, 7, 625-636, doi:10.5194/amt-7-625-2014, 2014.
- Shrivastava, M., Cappa, C.D., Fan, J., Goldstein, A.H., Guenther, A.B., Jimenez, J.L., Kuang, C., Laskin, A., Martin, S.T., Ng, N.L., Petaja, T., Pierce, J.R., Rasch, P.J., Roldin, P., Seinfeld, J.H., Shilling, J., Smith, J.N., Thornton, J.A., Volkamer, R., Wang, J., Worsnop, D.R., Zaveri, R.A., Zelenyuk, A., Zhang, Q., 2017. Recent advances in understanding secondary organic aerosol: Implications for global climate forcing. *Rev. Geophys.* 55, 509–559. <https://doi.org/10.1002/2016RG000540>
- Six et al., First aerosol optical thickness measurements at Dome C (East Antarctica), summer season 2003–2004, *Atmospheric Environment*, 39, 5041-5050, 2005.
- Shepherd, A., et al. (2018). Mass balance of the Antarctic Ice Sheet from 1992 to 2017. *Nature* 558, 219-222.
- Slusher, D.L., Neff, W.D., Kim, S., Huey, L.G., Wang, Y., Zeng, T., Tanner, D.J., Blake, D.R., Beyersdorf, A., Lefer, B.L., Crawford, J.H., Eisele, F.L., Mauldin, R.L., Kosciuch, E., Buhr, M.P., Wallace, H.W., Davis, D.D., 2010. Atmospheric chemistry results from the ANTCI 2005 Antarctic plateau airborne study. *J. Geophys. Res. Atmos.* 115, 1–18. <https://doi.org/10.1029/2009JD012605>
- Solomon, A., Feingold, G., and Shupe, M.: The role of ice nuclei recycling in the maintenance of cloud ice in Arctic mixed-phase stratocumulus, *Atmospheric Chemistry and Physics*, 15, 10 631–10 643, 2015.
- Solomon, A., G. de Boer, J. M. Creamean, A. McComiskey, M. D. Shupe, M. Maahn, and C. Cox (2018), The relative impact of cloud condensation nuclei and ice nucleating particle concentrations on phase partitioning in Arctic mixed-phase stratocumulus clouds, *Atmos. Chem. Phys.*, 18(23), 17047-17059, doi:10.5194/acp-18-17047-2018.
- Solomon, S.: Stratospheric ozone depletion: A review of concepts and history, *Rev. Geophys.*, 37, 275–316, <https://doi.org/10.1029/1999RG900008>, 1999.



- Souvereinjs, N., Gossart, A., Lhermitte, S., Gorodetskaya, I.V., Kneifel, S., Maahn, M., Bliven, F.L., van Lipzig, N.P.M. (2017). Estimating radar reflectivity - snowfall rate relationships and their uncertainties over Antarctica by combining disdrometer and radar observations. *Atmospheric Research*, 196, 211-223.
- Souvereinjs, N., Gossart, A., Gorodetskaya, I.V., Lhermitte, S., Mangold, A., Laffineur, Q., Delcloo, A., van Lipzig, N.P.M. (2018a). How does the ice sheet surface mass balance relate to snowfall? Insights from a ground-based precipitation radar in East Antarctica. *The Cryosphere* 12,1987-2003, 2018a
- Souvereinjs N., Gossart A., Lhermitte S., Gorodetskaya IV., Grazioli J., Berne A., Duran-Alarcon C., Boudevillain B., Genthon C., Scarchilli C., van Lipzig N.P.M. (2018b). Evaluation of the CloudSat surface snowfall product over Antarctica using ground-based precipitation radars. *The Cryosphere*, 12 (12), 3775-3789. doi: 10.5194/tc-12-3775-2018b.
- Souvereinjs, N., N. van Lipzig, (superv.), S. Lhermitte (co-superv.), I. Gorodetskaya (co-superv.) Precipitation and clouds over Antarctica from an observational and modelling perspective, Dissertation, Faculty of Science, Department of Earth and Environmental Sciences, Regional Climate Studies, Celestijnenlaan 200E, 3001 Leuven, Belgium, 255pp, 2019a.
- Souvereinjs, N., Gossart, A., Demuzere, M., Lenaerts, J. T. M., Medley, B., Gorodetskaya, I. V., Vanden Broucke, S., and van Lipzig, N. P. M.: A New Regional Climate Model for POLAR-CORDEX: Evaluation of a 30-Year Hindcast with COSMO-CLM2 Over Antarctica, *J. Geophys. Res.-Atmos.*, 124, 1405–1427, <https://doi.org/10.1029/2018JD028862>, 2019b.
- Spurr, R.: User's Guide VLIDORT Version 2.6, RT Solutions, Inc., 2013.
- Spurr, R.: VLIDORT: A linearized pseudo-spherical vector discrete ordinate radiative transfer code for forward model and retrieval studies in multilayer multiple scattering media, *J. Quant. Spectrosc. Ra.*, 102, 316–342, 2006.
- Spurr, R. J. D., Kurosu, T. P., and Chance, K. V.: A linearized discrete ordinate radiative transfer model for atmospheric remote-sensing retrieval, *J. Quant. Spectrosc. Ra.*, 68, 689–735, [https://doi.org/10.1016/S0022-4073\(00\)00055-8](https://doi.org/10.1016/S0022-4073(00)00055-8), 2001.
- Stohl, A., C. Forster, A. Frank, P. Seibert, and G. Wotawa (2005): Technical Note : The Lagrangian particle dispersion model FLEXPART version 6.2. *Atmos. Chem. Phys.* 5, 2461-2474.
- Stohl, A., P. Seibert, G. Wotawa, D. Arnold, J. F. Burkhart, S. Eckhardt, C. Tapia, A. Vargas, and T. J. Yasunari (2012): Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition. *Atmos. Chem. Phys.* 12, 2313-2343, doi:10.5194/acp-12-2313-2012.
- Tatzelt, C., Henning, S., Welti, A., Baccharini, A., Hartmann, M., Gysel-Beer, M., van Pinxteren, M., Modini, R. L., Schmale, J., and Stratmann, F.: Circum-Antarctic abundance and properties of CCN and INPs, *Atmos. Chem. Phys.*, 22, 9721–9745, <https://doi.org/10.5194/acp-22-9721-2022>, 2022.
- Teinilä, K., V.M. Kerminen and R. Hillamo, A study of size-segregated aerosol chemistry in the Antarctic atmosphere, *J. Geophys. Res.*, 105, 3893-3904, <https://doi.org/10.1029/1999jd901033>, 2000.
- Thalman, R. and R. Volkamer, Temperature dependent absorption cross-sections of O2-O2 collision pairs between 340 and 630 nm and at atmospherically relevant pressure., *Phys. Chem. Chem. Phys.*, 15(37), 15371–81, doi:10.1039/c3cp50968k, 2013.
- Tirpitz, J.-L., Frieß, U., Hendrick, F., Alberti, C., Allaart, M., Apituley, A., Bais, A., Beirle, S., Berkhout, S., Bogner, K., Bösch, T., Bruchkouski, I., Cede, A., Chan, K. L., den Hoed, M., Donner, S., Drosoglou, T., Fayt, C., Friedrich, M. M., Frumau, A., Gast, L., Gielen, C., Gomez-Martín, L., Hao, N., Hensen,

- A., Henzing, B., Hermans, C., Jin, J., Kreher, K., Kuhn, J., Lampel, J., Li, A., Liu, C., Liu, H., Ma, J., Merlaud, A., Peters, E., Pinardi, G., Pithers, A., Platt, U., Puentedura, O., Richter, A., Schmitt, S., Spinei, E., Stein Zweers, D., Strong, K., Swart, D., Tack, F., Tiefengraber, M., van der Hoff, R., van Roozendaal, M., Vlemmix, T., Vonk, J., Wagner, T., Wang, Y., Wang, Z., Wenig, M., Wiegner, M., Wittrock, F., Xie, P., Xing, C., Xu, J., Yela, M., Zhang, C., and Zhao, X.: Intercomparison of MAX-DOAS vertical profile retrieval algorithms: studies on field data from the CINDI-2 campaign, *Atmos. Meas. Tech.*, 14, 1–35, <https://doi.org/10.5194/amt-14-1-2021>, 2021.
- Tomasi et al., *Aerosol remote sensing in polar regions*, *Earth-Science Reviews*, 140, 108-157, 2015.
- Topping, D., P. Connolly, G. McFiggans (2013), Cloud droplet number enhanced by co-condensation of organic vapours, *Nature Geoscience*, 6, 443-446.
- Uotila, P., Vihma, T., Pezza, A.B., Simmonds, I., Keay, K. Lynch, A.H. (2011). Relationships between Antarctic cyclones and surface conditions as derived from high-resolution numerical weather prediction data. *Journal of Geophysical Research* 116, D07109.
- van den Broeke, M., Reijmer, C., van As, D., Boot, W. (2006). Daily cycle of the surface energy balance in Antarctica and the influence of clouds. *International Journal of Climatology* 26, 1587-1605.
- Van Tricht K., Gorodetskaya I., Lhermitte S., Turner D.D., Schween J., Van Lipzig N. (2014). An improved algorithm for polar cloud-base detection by ceilometer over the ice sheets. *Atmospheric Measurement Techniques*, 7, 1153-1167. doi: 10.5194/amt-7-1153-2014.
- Van Tricht, K., Lhermitte, S., Lenaerts, J.T.M., Gorodetskaya, I.V., L'Ecuyer, T.S., Noël, B., van den Broeke, M.R., Turner, D.D., van Lipzig, N.P.M. (2016). Clouds enhance Greenland ice sheet meltwater runoff. *Nature Communications* 7, 10266.
- Van Wessem, J.M. et al. (2018). Modelling the climate and surface mass balance of polar ice sheets using RACMO2 – Part 2: Antarctica (1979-2016). *The Cryosphere* 12, 1479-1498.
- Virkkula, A. et al., Review of aerosol research at the Finnish Antarctic research station Aboa and its surroundings in Queen Maud Land, Antarctica, *Geophysica*, 45, 163-181, 2009.
- Wagenbach, D. U. Görlach, K. Moser and K.O. Münnich, Coastal Antarctic aerosol: The seasonal pattern of its chemical composition and radionuclide content, *Tellus B*, 40, 426-436, <https://doi.org/10.3402/tellusb.v40i5.16010>, 1988.
- Wagner, T., Beirle, S., Benavent, N., Bösch, T., Chan, K. L., Donner, S., Dörner, S., Fayt, C., Frieß, U., García-Nieto, D., Gielen, C., González-Bartolome, D., Gomez, L., Hendrick, F., Henzing, B., Jin, J. L., Lampel, J., Ma, J., Mies, K., Navarro, M., Peters, E., Pinardi, G., Puentedura, O., Puķīte, J., Remmers, J., Richter, A., Saiz-Lopez, A., Shaiganfar, R., Sihler, H., Van Roozendaal, M., Wang, Y., and Yela, M.: Is a scaling factor required to obtain closure between measured and modelled atmospheric O<sub>4</sub> absorptions? An assessment of uncertainties of measurements and radiative transfer simulations for 2 selected days during the MAD-CAT campaign, *Atmos. Meas. Tech.*, 12, 2745–2817, <https://doi.org/10.5194/amt-12-2745-2019>, 2019.
- Walters, D., Boutle, I., Brooks, M., Melvin, T., Stratton, R., Vosper, S., Wells, H., Williams, K., Wood, N., Allen, T., Bushell, A., Copley, D., Earnshaw, P., Edwards, J., Gross, M., Hardiman, S., Harris, C., Heming, J., Klingaman, N., Levine, R., Manners, J., Martin, G., Milton, S., Mittermaier, M., Morcrette, C., Riddick, T., Roberts, M., Sanchez, C., Selwood, P., Stirling, A., Smith, C., Suri, D., Tennant, W., Vidale, P. L., Wilkinson, J., Willett, M., Woolnough, S., and Xavier, P.: The Met Office Unified Model Global Atmosphere 6.0/6.1 and JULES Global Land 6.0/6.1 configurations, *Geosci. Model Dev.*, 10, 1487–1520, <https://doi.org/10.5194/gmd-10-1487-2017>, 2017.

- Weller, R. and A. Lampert, Optical properties and sulphate scattering efficiency of boundary layer aerosol at coastal Neumayer station, Antarctica, *J. Geophys. Res.*, 113, D16208, <https://doi.org/10.1029/2008JD009962>, 2008.
- Weller, R., K. Schmidt, K. Teinilä and R. Hillamo, Natural new particle formation at the coastal Antarctic site Neumayer, *Atmos. Chem. Phys.*, 15, 11399-11410, <https://doi.org/10.5194/acp-15-11399-2015>, 2015.
- Wex, H., L. Huang, W. Zhang, H. Hung, R. Traversi, S. Becagli, R. J. Sheesley, C. E. Moffett, T. E. Barrett, R. Bossi, H. Skov, A. Hünerbein, J. Lubitz, M. Löffler, O. Linke, M. Hartmann, P. Herenz, and F. Stratmann (2019), Annual variability of ice nucleating particle concentrations at different Arctic locations, *Atmos. Chem. Phys.*, 19, 5293–5311, doi:10.5194/acp-19-5293-2019.
- Woolfenden, E., Cole, A., 2003. Sampling Device - US 6564656 B1. 6564656.
- Woolfenden, E., 2012. Thermal Desorption for Gas Chromatography, *Gas Chromatography*. <https://doi.org/10.1016/B978-0-12-385540-4.00010-9>
- Wood, N.B., L'Ecuyer, T.S., Bliven, F.L., Stephens, G.L. (2013). Characterisation of video disdrometer uncertainties and impacts on estimates of snowfall rate and radar reflectivity. *Atmospheric Measurement Techniques* 6, 3635-3648.

## ANNEXES

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- 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](https://doi.org/10.1007/978-3-030-52171-4_18)

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- Mottram, R., Hansen, N., Kittel, C., van Wessem, J. M., Agosta, C., Amory, C., Boberg, F., van de Berg, W. J., Fettweis, X., Gossart, A., van Lipzig, N. P. M., van Meijgaard, E., Orr, A., Phillips, T., Webster, S., Simonsen, S. B., and Souverijns, N.: What is the surface mass balance of Antarctica? An intercomparison of regional climate model estimates, *The Cryosphere*, 15, 3751–3784, <https://doi.org/10.5194/tc-15-3751-2021>, 2021