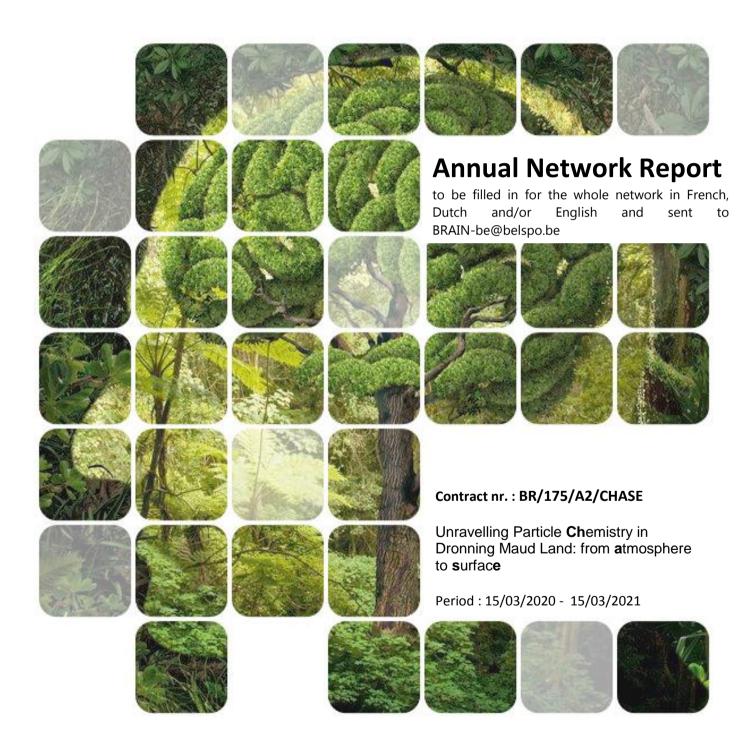


BRAIN-be

BELGIAN RESEARCH ACTION THROUGH INTERDISCIPLINARY NETWORKS



NETWORK

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PROJECT WEBSITE:

https://ozone.meteo.be/projects/chase

Yearly, one report (max. 15-20 pages) should be filled in for the whole network in French, Dutch or English and sent to BRAIN-be@belspo.be.

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1. EXECUTIVE SUMMARY OF THE REPORT

The CHASE project provides detailed physical-chemical analyses of both atmospheric and surface snow particles as well as of (volatile) organic compounds recovered near the Belgian research station Princess Elisabeth (PE), Dronning Maud Land, East Antarctica, and thoroughly investigates their atmospheric transport pathways. Such detailed studies have never occurred in the region where Princess Elisabeth station is located. The project consists of 4 components: (i) a particle and air sampling with physical-chemical analysis component, (ii) a data interpretation component, (iii) a synthesis component, and (iv) a valorisation component.

The work is subdivided in several tasks and deliverables, executed by the different partners of this project. Their progress regarding the different deliverables is listed in table 1. The start date of the project was the 1^{st} January 2017. In the table below, the submission date is counted from the 15^{th} April 2017 (48 months until end of project in contract 15/04/2021 – however, prolonged to 15/04/2022).

No.	Description	Partner	Subm. date	Status
D1.1	Active and passive sampling methods for the atmospheric organic composition analysis of both particulate and gaseous fraction	UGent	M12, M24, M36, M46	FIN
D1.2	Advanced analytical procedures enabling detailed molecular characterization of collected air samples by using highly innovative mass spectrometry based equipment	UGent	M12, M24, M36, M46	PROG
D1.3	Unique dataset on detection frequencies and concentration levels of organic micropollutants in both Austral Summer and Winter at Dronning Maud Land	UGent	M12, M24, M36, M48	PROG
D1.4	Analysis methods developed for stable isotopes C and N of the organic aerosol fraction and related dataset on isotopic composition of the organic fraction of particulate matter	VUB	M12, M24, M36, M48	PROG
D2.1	Active sampling and analysis methods developed for inorganic composition of atmospheric particles and related dataset of inorganic composition	ULB	M12, M24, M36, M48	PROG
D2.2	Passive sampling and analysis methods developed for inorganic composition of atmospheric particles and related dataset of inorganic composition	ULB M12, M24, M36, M48		PROG
D2.3	Surface snow samples collected and analysis methods developed for inorganic composition of particles therein and related dataset of inorganic composition	ULB M12, M24, M36, M48		PROG
D3.1	Air mass trajectories calculated, dispersion analysis of atmospheric pathways, clustering of source regions	RMI	M12, M24, M36, M42	PROG
D4.1	Source regions, transport pathways, seasonal variations and relative importance of trace elements, micronutrients and atmospheric pollutants, of natural and anthropogenic compounds	RMI	M18, M30, M48	PROG
D4.2	Cloud Condensation Nuclei and Ice Nuclei characterisation	RMI	M18, M30, M48	PROG
D5.1	Management of the network	RMI	Cont.	PROG
D5.2	Quality controlled chemistry database	RMI	Cont.	PROG
D5.3	Results published to scientific community, policy and public	RMI, UGent, ULB, VUB	Cont.	PROG
D5.4	Scientific workshop	RMI, UGent, ULB, VUB	M42.	NOT

Table 1: List of intermediate and final deliverables and their dissemination. The first three columns give the number, the description and the partner responsible for the deliverable, the fourth column gives the submission date, counted from 15 April 2017, and the fifth column gives the status (finished (FIN), in progress (PROG), or not started (NOT)).

After the end of the BELARE season 2019/20 in February 2020, the samples arrived, due to the Covid-19 pandemic and related restrictions, only by June 2020 at the partner's institutes and were stored in freezers and clean rooms for further analyses.

With respect to the analyses of (volatile) organic compounds, it can be said that the obtained results are promising, and the developed methods prove to be robust and enable to compare data between the different sample years. Certainly, a unique dataset will be created containing a broad range of detected compounds and concentration levels.

The first results obtained by the Lab G-Time confirm the importance of dust source areas, like the South of Africa (SAF), underestimated until now. SAF is revealed as a major contributor for atmospheric depositions in Antarctica nowadays and in the past (Last Glacial Maximum). In addition to the SEM-EDS results, A. Vanderstraeten developed a novel statistical model of rare earth pattern compilation providing a sensitive and powerful tool for high resolution identification of the dust source area contributions reaching East Antarctica. This innovative model can be applied for the Southern Hemisphere and the Northern Hemisphere as well (Greenland), and for the Glacial or Interglacial past periods in polar regions. It is a key advance for a better knowledge of the modern climate system and for the understanding process of the past and future global changes.

In collaboration with the Glaciology and G-Time groups from ULB, the AMGC group (VUB) studied the surface snow samples (from 2017 and 2019) for major ion analysis, organic matter concentrations and C isotopes (i.e., particulate and dissolved fractions), covering a gradient from the Antarctic Plateau to the coast while passing through the vicinity of the Belgian research station Princess Elisabeth, Dronning Maud Land, East Antarctica. Major ion analysis (Na+, K+, Mg2+, NH4+, Cl-, SO42-, NO3-, and methanesulfonic acid) will provide insights on the contribution of the ocean to the aerosols in this region, with expected gradients from the coast to the plateau.

The FLEXTRA trajectory model has been applied in order to investigate possible source regions and transport pathways into Antarctica of atmospheric particles and SVOCs. 10-days backward trajectories, starting from PEA, were calculated for a 10 year period. A k-means cluster analysis has been done and four clusters of air mass origin were found. The cluster analysis has been done for the whole period and also for each season separately. Some distinct features can be seen in the air mass origin clustering. Within the constraints of this analysis, 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.

The following preparations were undertaken for the 2020/21 field campaign at Princess Elisabeth Antarctica (PEA) station:

- Virtual meetings and email-exchanges with the Station Operator in order to discuss the practical topics for the sampling campaign;
- Preparation of the necessary air cargo boxes and shipment forms;
- Administrative organization of the campaign in cooperation with the polar secretariat and the station operator;
- Material for the filter sampling has been prepared and ordered. Cleaning and preparation of the bottles needed for sampling surface snow;
- Preparation of the aethalometer instrument for re-installation at PEA after repair in Belgium

Preben Van Overmeiren participated on the CHASE project in the BELARE 2020/2021 field campaign to PEA station. He was accompanied by Alexis Merlaud from the Royal Belgian Institute for Space Aeronomy (IASB-BIRA), who was there for the closely related CLIMB project. Preben Van Overmeiren stayed from end of November 2020 to mid-January 2021 at PEA. The scientists re-installed the active sampling instrumentation in the northern 'Atmos' shelter of PEA. At the sites for passive sampling (see list below and Fig. 1), samples of the last winter period were collected

and new samples for atmospheric particles and volatile organic compounds installed. At two of the passive sampling sites (Frank Kenny North and Plateau) 3-m long ice cores for chemical analyse were successfully collected. At six sites, also surface snow samples were collected. The collected samples should arrive by end of April 2021 in Belgium and will be stored either at Ghent University or at Laboratoire G-Time (ULB) for further analyses. Alexis Merlaud and Preben Van Overmeiren helped in addition to supervise instrumentation for atmospheric aerosol physical properties, cloud and precipitation monitoring, helped with launching radio sondes, and took care also of tasks of the CLIMB project.

Further details are described below within the progress per task section.

Coordinates and altitude (m. asl.) of the sites for passive and surface snow sampling:

			- · · · · · · · · · · · · · · ·		
٠	East of PEA station:	71.96014 °S	23.47353 °E	1320 m	operational, @, **, §
٠	Plateau:	72.25336 °S	23.23195 °E	2300 m	operational, @, **, §
٠	Deep Plateau:	72.37655 °S	23.41896 °E	2370 m	operational, @, **
٠	Romnoes:	71.34678 °S	23.61131 °E	700 m	@, #,
٠	Frank Kenny South:	70.82900 °S	23.73500 °E	320 m	@, **, #
٠	Frank Kenny North:	70.43281 °S	23.84089 °E	110 m	operational, @, **
٠	Breid Bay (coast):	70.30485 °S	23.61642 °E	75 m	#, **

@ passive samples collected

passive sampling de-installed

- ** snow samples collected
- § site moved: PEA 500m further; Plateau to CLIMB site (72.27101 °S, 23.25238 °E, 2350 m)

Figure 1 shows the locations on a map of Dronning Maud Land.

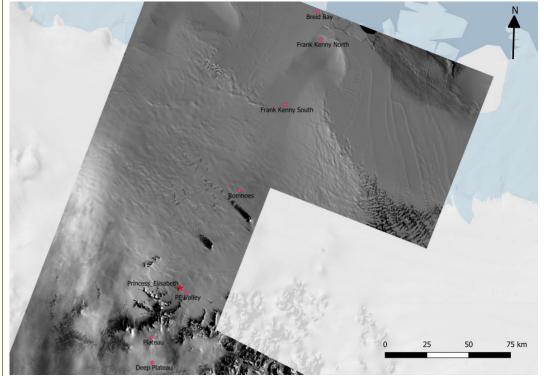


Figure 1: Location of the seven sampling sites where the CHASE passive samplers have been installed and where surface snow samples have been collected. 'Deep plateau', southward of PE, on the plateau; ii) 'Plateau', southward of PE, vicinity of the plateau; iii) 'PE valley', around 4 km eastward of PE;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

2. ACHIEVED WORK

Detailed description of the achieved work and tasks of the past reporting year

Task 1: Characterisation of the organic atmospheric composition (particulate matter and VOCs) (UGent, VUB)

Task 1.1: Sampling and sample preparation of atmospheric particles for organic analysis (UGent)

The Digitel DHA-80 High Volume Sampler (HVS, 500 l/min) for active sampling of atmospheric particles has been re-installed in a specific container around 300 m north of PE station. The active sampling is limited to the austral summer period (filter exchange, energy demand). Pre-baked quartz-fibre filters have been used for the collection of particulate matter together with polyurethane foam filter cartridges to capture more volatile components. During the 2020-2021 austral summer the instrument ran for the 4th consecutive year, creating a unique time resolved sample set of trace organic chemicals in the East-Antarctic air and particulate phase.

Simultaneously with the high volume active sampling, polyurethane foam disk passive samplers have been installed, to be able to identify trace organic semi-volatile and non-volatile micro-pollutants. Samplers were initially installed at 5 sites in the 2017-2018 season. In the 2018-2019 season the experimental capacity was expanded with the two extra sites and a second sample set was acquired in season 2019-20 from the first 5 sites and a first sample set from the other two sites. During the 2020-2021 season all seven sites were visited for sample collection and filter replacement or removal of sampling poles. 2 polyurethane disk passive samplers were installed on the roof of the North shelter as reference for extra quality control.

All instrumentation on the removed passive sample sites mentioned in section 1 was successfully recovered and re-used or sent back to Belgium.

Task 1.2: Sampling and sample preparation for the analysis of Volatile Organic Compounds (UGent)

Volatile Organic Compounds (VOCs) were sampled by passive sampling. Axial passive samplers have been installed on poles (around 2-3 m above ground) at the same locations as the passive samplers for semi-volatile organic analysis mentioned before. In November, December 2020 the samples were collected and new ones were installed. These samplers will collect VOCs over another year until final recovery in November, December 2021.

An in-house developed active sampler was installed on the roof of a scientific shelter close to the station. This instrument sequentially takes a new sample every 2 weeks for 1 year, so a time resolved series, covering also the Antarctic winter will be obtained. This will give insight how the organic load in the atmosphere changes over different seasons. Breakthrough tests verified that no breakthrough occurred for the compounds of interest with the pre-set sample volume. The latter instrument is now maintained in the framework of the CLIMB project.

All instrumentation on the removed passive sample sites mentioned in section 1 was successfully recovered and re-used or sent back to Belgium.

Task 1.3.1: Laboratory analysis for the molecular characterisation of the organic atmospheric composition (UGent)

From the past 3 sample seasons all high-volume samples, both the PU Foams as well as the QFFs, are successfully extracted (PLE) and analysed for both PAH's and OPAH's (GC-HRMS). Several species of both groups are found in quantifiable amounts. New samples arriving in Q2 of 2021 can be analysed as soon they are available.

The extraction of the passive PUFs was impeded by the co-extraction of a large amount of matrix. A new method for cleaning up the extracts is being developed before continuing the extraction of

the remaining passive samples. A clean-up step using a Florisil SPE cartridge with an additional layer of drying agent seems promising but is yet to be validated using surrogate samples which were taken in during the 20-21 expedition and will be spiked with the targeted compounds. Sorbent tubes collected during the 19-20 expedition were analysed on thermal desorption GC-MS and TD-PTR-TOFMS using the already developed and validated method. The samples collected during the 20-21 season will be analysed as soon as they become available and will mark the third consecutive year in this unique data set.

Task 1.3.2: Laboratory analysis for the stable isotopes C and N of the organic aerosol fraction (VUB)

For carbon content and isotope data, a first snow sample from the coast was analyzed by the VUB lab (after filtration performed at the G-Time clean lab). On the basis of these preliminary results obtained at the AMGC lab (VUB), a master project was proposed to a student (Femke Buys from Gent University) under the supervision of Prof. Philippe Claeys and François Fripiat for the 2020-21 academic year. The goal of this master thesis is to measure the C-content and isotopic composition (in particulate and dissolved fractions), and major ions (Na+, K+, Mg2+, NH4+, Cl-, SO42-, NO3-, and methanesulfonic acid) in the snow samples taken during the field campaigns in 2017 (Frank Kenny North and South and Princess Elisabeth station) and 2019 (Breid Bay, Frank Kenny North and South, Romnoes, Princess Elisabeth station, Plateau and Deep plateau), along the coast-inland transect to untangle the marine aerosol contribution relative to the continental contributions.

Task 1.4: Interpretation of the results for organic atmospheric composition (UGent, VUB)

UGent is investigating together with partner RMI if the trends observed in the PAH/OPAH dataset are correlated with meteorological data.

Within the scope of the Master thesis of Femke Buys, the C elemental and isotopic data are compared with the major ion abundance data all along the transect defined from the inland sites (plateau and deep plateau) to the coastline. The data are compiled according to the snow accumulation and sea salt contents. The marine contributions and hopefully the anthropogenic contributions should be revealed along this transect.

Task 2: Characterisation of the inorganic composition of atmospheric particles (ULB)

Task 2.1: Active sampling and analysis of inorganic composition of atmospheric particles (ULB)

Active sampling using 0.2 μ m pore-size Teflon filters has been done from December 2020 (CHASE team) through January 2021 (PE station staff). For this active sampling, the same system as the previous campaign was used: a strong pump (nominal flow rate of 330 L/min) was re- installed in the same "atmosphere" container, together with the re-installation of the inlet on the roof of the container. A flow meter was also re-installed in the sampling line in order to derive accurate values for actual and total sample flows. In addition to several blank samples, 3-4 filter samples were recovered. These filter samples are already in Belgium and awaiting laboratory analysis.

However, a malfunctioning of the pump lead to a limited number of samples this year compared to the previous campaigns. However, these filters still guarantee the follow-up of the temporal records throughout several years of sampling.

SEM-EDS analyses of the filters from the previous and 2020 campaigns are planned to be performed by Sibylle Boxho (teaching assistant and PhD student since October 2020) at LPCA – ULCO, Université Côte d'Opale, Dunkirk – France. SEM-EDS are the abbreviations of a Field Emission Scanning Electron Microscope (i.e., SEM)(JeoITM JSM-7100F) equipped with three energy dispersive X-ray detectors (EDS)(Bruker XFlash 6/30) hosted by the laboratory of LPCA – ULCO. Single particle analyses will be performed both in manual or automatic modes.

Task 2.2: Passive sampling and analysis of inorganic composition of atmospheric particles (ULB)

During the season 2020-2021, six sampling sites were visited and the Sigma-2 passive collectors were opened for the recovery of the Teflon filter (sample). A new clean filter was left in place at each of the remaining sites (see section 1) for the collection of inorganic particles over another year until the next campaign. A savilex® beaker was also added next to the filter holder for collection of the atmospheric fallouts, for testing another way of dust sampling, in order to reduce the risk of fine particle loss due to potential resuspension or perturbations by snow accumulation inside the sigma 2 (e.g., during storms).

SEM-EDS analyses of the Sigma 2 dust filters from the previous campaigns were planned by Aubry Vanderstraeten and Stefania Gili. They were performed on a Field Emission Scanning Electron Microscope (i.e., SEM)(JeoITM JSM-7100F) at LPCA – ULCO, Université Côte d'Opale, Dunkirk – France, which is equipped with three energy dispersive X-ray detectors (EDS)(Bruker XFlash 6/30 with 30 mm² polymer ultrathin-windows each), enabling the analysis of elements with an atomic number higher than boron ($Z \ge 5$).

Task 2.3: Sampling of surface snow and analysis of inorganic composition of particles therein (ULB)

Surface snow samples have been taken in November and December 2020 at 6 locations: i) around 4 km eastward of PE station; ii) one site southward of PE, in the vicinity of the plateau; iii) 16 km from that site, a new site at the deep plateau; iv) in between Romnoes and the coast; v) at one site at the western part of an ice rise at the coast and, vi) 'Breid Bay', near the coastline, Breid Bay. A total of 30 bottles of 10 L, i.e., a total of ~300 L of surface snow, have been collected. The bottles have been shipped back and were delivered in our lab on April 23, where they will be treated for chemical and isotopic analyses of the particles from the filtered snow samples in ultra-clean laboratory conditions. In addition, at the Frank Kenny north and the Plateau sites, 3-m ice cores were drilled for the analysis of inorganic particles.

To isolate the dust fraction, the 2019 snow samples (as for the previous campaign samples) were filtrated under flow hood in the clean lab at the G-Time. The dust samples were then dissolved in a (HF+HNO₃+HCl) mixture in Savillex® vials before being prepared for rare earth (REE) concentration analyses (by ICP-MS Agilent 7700 at G-Time and HR-ICP-MS Element 2 at the VUB) and Sr, Nd and Pb isotopic measurements on a Multi Collector ICP Mass Spectrometer (MC-ICP-MS) Nu Plasma II in dry mode (DSN desolvating system).

A novel statistical model was developed to compile the REE profiles of the Antarctica dust samples with those from all related dust PSAs (potential source areas) from the southern hemisphere. This innovative approach should help to discriminate and quantify the PSA respective contributions to Antarctica atmospheric fallouts. It should also provide a complementary powerful tool to the isotopic tracing method for identification of the origin of the mineral dust, especially when the sample amount is a limiting factor.

Task 2.4: Interpretation of the results for inorganic particle composition (ULB)

For atmospheric particle depositions on filters collected by the Passive Sigma2 collector or the active sampling system (installed on the Atmosphere container near the polar station), SEM-EDS analyses in automatic mode (mapping) and manual mode (for measurements of more precise semiquantitative chemical composition per single particle) have been done and are still planned. The compilation of this original SEM-EDS dataset provides grain- size, shape and semi-quantitative chemical analysis of several thousand atmospheric particles from six key samples collected along a transect perpendicular to the coastline. In addition to cluster statistical interpretation from the high amount of particle analysed, SEM-EDS analysis also gives the opportunity to detect specific external or internal assemblages or mixtures of particles. Initially, clustering statistical analysis was applied to distinguish particle types and better understand the variability within the whole sample sets. However, only mineralogical trends can be obtained this way. Therefore, to get further detailed mineralogical identification, we developed a Matlab script to classify each particle in a specific mineralogical group, which is based on the particle elemental composition. These analyses will provide a better understanding of the type of particle reaching the Antarctic coast to better constrain their origin.

Over the last few decades, it has been demonstrated the main dust contributor to the SO and Antarctica is mainly southern South America (SSA). Secondary contributions from other Potential Source Areas (PSAs) were also suggested, such as Australia (AUS), New Zealand (NZ), Southern Africa (SAF) and a local Antarctic source. Revisiting the literature, it became apparent that there is still no consensus which of these PSAs contribute to dust deposition over Antarctica -beyond the SSA source- mainly due to the lack of systematic studies of specific PSAs and the very small quantity of dust that is actually deposited over Antarctica, which leads to analytical challenges for geochemical studies. While most of the studies have been focused on the detailed study of SSA and AUS dust sources, other PSAs, such as SAF, are a clear example of the need for more extended geochemical studies to fill the gap in understanding the past and present long-range transportation of mineral dust particles to the southern hemisphere atmosphere. The Namib Desert coast was only recently identified as one of the largest sources of dust in Southern Africa, Within the Namib Desert and along the coast, the Kuiseb, Omaruru and Huab Riverbeds are the three main areas of greatest dust emission located on recently deposited fluvial surfaces. Therefore, in addition to the preparations and analyses of the Antarctic snow samples, preparations and analyses for REE concentrations and Sr, Nd and Pb isotopic compositions were also carried out on bulk and fine fractions of South Africa sediment samples. Sediment samples were dissolved in a (HF+HNO3+HCI) mixture in Savillex® vials at the G-Time Lab, Université Libre de Bruxelles. REE concentrations were analyzed by HR-ICP-MS Element 2 at the VUB. Strontium-Nd-Pb were chemically separated through ion-exchange resins following the procedure reported in (Vandersraeten, A et al., Geostandards and Geoanalytical Research, doi:10.1111/ggr.12320) and their isotopic compositions were analyzed using the MC-ICP-MS Nu Plasma II in dry mode at ULB. In addition to the 13 Antarctic snow samples, 22 bulk SAF samples (9 samples from Huab, 7 from Omaruru and 6 from Kuiseb) were analyzed for their REE and Sr-Nd-Pb isotope compositions. In order to evaluate a possible change in the geochemical fingerprint due to grain-size effects, 6 bulk samples from the Huab valley were selected to generate the fine fraction (< 5 μ m) using an atmospheric simulation chamber (in collaboration with Paola Formenti - CESAM analytical platform, Paris - France).

To further specify the geochemical signature of the local rocks, also potential dust precursors, five rocks and two cryoconite samples were collected and analyzed for their REE patterns and Sr, Nd and Pb isotopes at G-Time laboratory. The novel statistical model was applied on the REE patterns of Antarctica dust samples, rocky samples, and those from all related PSAs to discriminate and quantify their potential respective contributions.

Task 3: Air mass tracing by dispersion analysis of atmospheric transport (RMI)

Task 3.1: Calculation of air mass trajectories (RMI)

The FLEXTRA trajectory model has been applied in order to investigate possible source regions and transport pathways into Antarctica of atmospheric particles and SVOCs. The model was driven with ECMWF meteorological fields. 10-days backward trajectories, starting from PEA, were calculated for the period 01/01/2010 to 31/12/2019, in 6-hour-intervals (the period will be prolonged to include also 2020). A k-means cluster analysis has been done based on several parameters. When the clustering is performed relying on the normalised latitude, longitude and altitude, four clusters of air mass origin are found.

Task 4: Implications of the found results for atmospheric transport of trace elements, micronutrients and pollutants towards Antarctica and its closely associated Southern Ocean (RMI)

Task 4.1: Trace elements, micronutrients and atmospheric pollutants in Antarctica – their source regions, transport pathways, seasonal variations and relative importance of natural and anthropogenic compounds (RMI)

Air mass origin analyses have been done in order to investigate if the found source areas support the chemical fingerprint of particles found in certain snow samples, pointing to distinct source areas in Southern Africa or Southern South America, respectively. However, with an improved statistical model, which ULB partners applied to the data of rare-earth elements from snow samples, the local rocks of the Sor Rondane mountains at PEA cover the entire set of fingerprint data from Southern America and the South of Africa. Therefore, new rock samples and snow samples have been collected during the field campaign 2020/21, in order to help to discriminate local from distal contributions.

Task 4.2: Implications of found particle chemistry on cloud condensation and ice nuclei (RMI)

Measured atmospheric aerosol properties at PES (total particle number, optical properties, size) have been further analysed in order to link them to the number of cloud condensation nuclei, based on the paper by Herenz et al (2019; <u>https://www.atmos-chem-phys.net/19/275/2019/</u>). Based on the INP samples of seasons 2017/18 and 2018/19, the sample time for INP filter collection was kept to 10 days for the campaign 2020/21. A total of 6 samples, plus blank samples, could be collected.

Task 5 Coordination, database management and valorisation (RMI, UGent, ULB, VUB)

Task 5.1: Network management (RMI)

Project coordination is led by the Royal Meteorological Institute. Several virtual meetings (due to Covid-19 travel and gathering restrictions) of partners took place before and also during the BELARE 2020/21 campaign, in order to prepare the campaign, discuss analysis progress and progress of the campaign. A general meeting of Chase partners took place on 26 October 2020 via an online meeting. On 9 March 2021, there has been a debriefing online meeting.

Task 5.2: Management of the chemistry database (RMI)

The database has not started yet. It is more reasonable to wait for a more complete dataset and to wait that results got published We will however publish on the website a detailed description which kind of measurements have been done at each sample location. The website can be found at https://ozone.meteo.be/projects/chase

Task 5.3: Publication of results to the scientific community, policy stakeholders and the general public (RMI, UGent, ULB, VUB)

CHASE partners submitted papers to *Atmospheric Chemistry and Physics*, *Geostandards and Geoanalytical Research*, and *Nature Communications*.

Manuscripts are planned to be submitted to *Nature: communications earth & environment*, *Science Advances* (AAAS); and *Geophysical Research Letters*.

CHASE partners presented results on several international scientific conferences. The general public has been addressed at several occasions and blogs have been maintained during the field campaign by Preben Van Overmeiren and by Alexander Mangold. For a detailed list, please see section 7.

Task 5.4: Scientific workshop (RMI, UGent, ULB, VUB) Not started yet.

3. INTERMEDIARY RESULTS

Task 1.1: Sampling and sample preparation of atmospheric particles for organic analysis (UGent)

All PUFs sampled using the high-volume sampler were extracted following the developed protocols for the PLE and analysed for PAHs and OPAHs using the validated method on GC-HRMS.

The extraction parameters for the QFFs was optimized and validated using urban dust reference material (NIST SRM 1649B) to account for matrix effects. Recoveries between 60 and 100% are now obtained while maintaining low blank levels.

Two clean-up methods were evaluated and compared to remove the co-extracted matrix from the extracts of the passive PUF samples. In a first study blank and recovery levels were checked for 2 types of SPE columns, Supelco EZ-POP NP and Florisil, with the latter providing better quality parameters such as earlier elution and high recoveries as well as being compatible with the existing GC-MS method. The clean-up method will be validated using spiked surrogate samples.

Task 1.2: Sampling and sample preparation for the analysis of Volatile Organic Compounds (UGent)

Samples obtained from the 19-20 season were analysed on GC-MS using the developed method to dry the samples before thermal desorption. The possibilities of recollection of samples for reanalysis are yet to be evaluated. TD-PTR-TOFMS offers an interesting new insight into the compounds absorbed in the sorbent tubes and is complementary to GC-MS but data interpretation remains tedious. This technique will be further explored.

Task 1.3.1: Laboratory analysis for the molecular characterisation of the organic atmospheric composition (UGent)

In all samples from the past three campaigns PAHs with a lower molecular weight (Acenaphthene to Chrysene – 3 to 4 rings) are detected in the PU foam filter indicating their presence in the gas phase. Total PAH concentrations derived from the PUFs varies between 5 and 100 pg/m³, with pyrene, fluorene, phenanthrene and fluoranthene being the main contributors. Remarkably also OPAHs were detected in concentration levels in the same order of magnitude as the PAHs, indicating the PAHs in the air masses could be largely transformed during atmospheric transport. A significant decreasing trend is found between the OPAH/PAH ratio and the total PAH concentration (see Fig. 2), suggesting high PAH concentrations are representing an air mass with a shorter transport time. This hypothesis is to be validated using back-trajectory analysis.

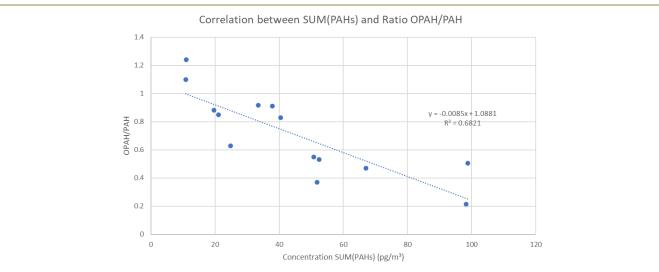


Figure 2: correlation between the total PAH concentration and the ratio of OPAH/PAH

The data of the QFF corresponding with particle-bound PAHs is still to be analysed. Data of the VOC measurement from the 19-20 season is yet to receive the same data treatment as the samples from the previous season.

Task 1.3.2: Laboratory analysis for the stable isotopes C and N of the organic aerosol fraction (VUB)

Surface snow has been sampled for major ion analysis, organic matter concentrations and isotopes (i.e., particulate and dissolved fractions), covering a gradient from the Antarctic Plateau to the coast while passing through the vicinity of the Belgian research station Princess Elisabeth, Donning Maud Land, East Antarctica. The analyses are being performed but preliminary data highlight a relatively constant isotopic composition for particulate organic carbon along the sampling gradient, with an isotopic signature being characteristic of the C3 plants of terrestrial origins.

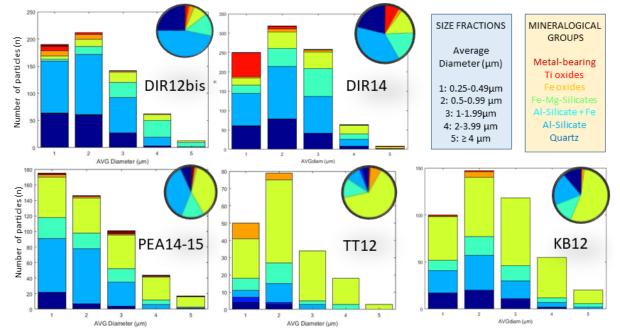
Clear concentration gradients are observed for the concentration of particulate organic carbon, i.e., increasing inland. Further analysis of the surface mass balance (i.e., net accumulation of snow) for a given month at the station will allow us to test if the observed gradients are due to a dilution effect by the amount of snowfall or due to variable concentrations of organic aerosols in the atmosphere at a given location. Major ion analysis (Na+, K+, Mg2+, NH4+, Cl-, SO42-, NO3-, and methanesulfonic acid) will provide insights on the contribution of the ocean to the aerosols in this region, with expected gradients from the coast to the plateau.

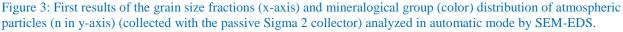
Task 2.1: Active sampling and analysis of inorganic composition of atmospheric particles (ULB)

The compilation of the original SEM-EDS dataset obtained for airborne particles collected on filters by active sampling – as for the passive sampling system, should provide grain- size, shape and semi-quantitative chemical analysis of several thousand atmospheric particles. In addition, SEM-EDS analysis also gives the opportunity to detect specific external or internal assemblages or mixtures of particles. The first analyses performed on filters from the Atmosphere container located nearby the polar station were realized with a simple polarized microscope, which confirm the significant higher quantity of particles on the filters. SEM-EDS analyses in automatic modes show preliminary results consistent with observations obtained from the Sigma-2 filters, confirming the mineralogy of the dust particles.

Task 2.2: Passive sampling and analysis of inorganic composition of atmospheric particles (ULB)

Based on the filters collected with the passive Sigma-2 sampling system, the SEM-EDS results highlight the very small particle size (< 5µm) of all samples, even near the Sør Rondane mountains and a mineral transect suggesting an influence from local rocks in addition to distal inputs at the coast. Mineralogical identification of individual particles can be based on their elemental composition obtained by automated analysis. Initially, clustering statistical analysis was applied to distinguish particle types and better understand the variability within the whole sample sets. However, only mineralogical trends can be obtained this way. Therefore, for detailed mineralogical identification, the Matlab script was used to classify each particle in a specific mineralogical group, which is based on the particle elemental composition. Among all particles analysed, aluminosilicate, silica, titanium and iron oxides particles are identified as "pure" particles according to their elemental composition. However, even though all these particles are considered as pure particles, they often contain low fractions of unexpected elements, like iron, aluminium and silicon.





Task 2.3: Sampling of surface snow and analysis of inorganic composition of particles therein (ULB)

In this study, 13 snow samples collected at 9 sites along a 250 km transect from the coastline to the inland Sør Rondane mountains (SR) in NE Antarctica were analysed for REE and SR, Nd and Pb isotopic compositions. This transect covers the transition between cyclonic (coastal) and anticyclonic (interior) weather systems, as well as the transition zone where both systems alternate. So far, most of the efforts to trace dust from ice cores have been focused on the central eastern regions of Antarctica, while the provenance of dust reaching the NE Antarctica coast remains poorly known although it probably better reflects the atmospheric fallouts into this sector of the Southern Ocean (i.e., Riiser-Larsen sea).

The Sør Rondane mountains are located parallel to the coastline ~200 km inland and are mainly exposed to katabatic winds from central Antarctica, making the SR an important source of local dust. Five rocks and two cryoconite samples were collected and analysed for their REE patterns and isotopic compositions. Near the Antarctic coastline, the atmospheric circulation is dominated by synoptic winds, which can transport dust from distal sources across the SH. Thus, we compiled

from literature the REE patterns of all PSAs. In addition, we analysed the REE signature of 18 sediment samples collected in three coastal basins of Namibia, SAF (Huab, Omaruru and Kuiseb), which were recently identified as major dust source regions to South Atlantic Ocean and possibly Southern Ocean. Sediments of those three basins were re-suspended in a dust chamber from which the airborne fraction was collected for analysis.

Our novel statistical approach compiles the REE patterns from 31 potential source areas (PSAs) which are statistically evaluated against 13 modern dust samples recovered along a 250 km transect in North-East Antarctica, from PEA to the coastline. Correlation coefficient (R), Akaike and Bayesian Information Criterion (AIC-BIC) are used to determine whether a dust sample traces a single or multiple PSAs. Our analyses indicate that local sources control the dust in the inland section of the transect, while distal sources dominate the coastal sites. Distal source areas include the previously identified Puna Altiplano, Patagonia but also Southern Africa, which is identified for the first time in recent dust in NE Antarctica. Our novel REE approach compiling 31 PSAs all over the Southern Hemisphere offers an effective and cost-efficient way to trace dust sources of Antarctica in the present and/or in the past.

In order to assess the influence of the Namibia region as a dust source to the high latitude environments of the southern hemisphere, we compare the SAF dust isotopic and geochemical signature with the dust fingerprint for the modern snow samples from Dronning Maud Land. For comparison purposes, we also included a re-analysis of the isotopic composition of SAF samples presented in previous studies. Due to the limited number of samples, the current characterization of SAF dust sources is poor and, as a consequence, the role of SAF as a dust contributor to the SO and Antarctica has plausibly been overlooked. Southern Africa appears as another major dust source contributor for Antarctica, besides Southern South America, at least during modern periods (Fig. 4). In addition, we reported the dust isotopic fingerprints for Antarctic ice cores (Vostok and EDC) during interglacial periods (Holocene and MIS 5.5) and compared those with the PSAs isotopic fields. Again, our results show that together with the Puna-Atiplano Plateau in SSA, the Huab, Omaruru, Kuiseb, and Namib sand sea in SAF, are significant dust sources and have a dust signature found in marine and ice cores during the Holocene (Fig. 5).

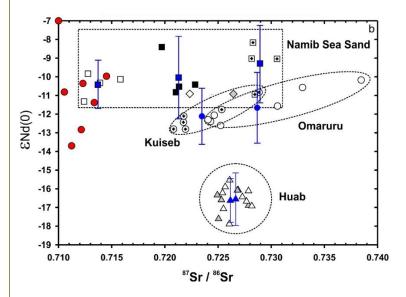


Fig. 4: Sr and Nd isotopic data for the modern dust collected in snow samples from our study transect in East Antarctica (red dots) and the signature of four dust source areas in Southern Africa (Huab, Omaruru, Kuiseb and Namibia Sea Sand). The blue lines represent the 2-sigma error bars for Nd composition.

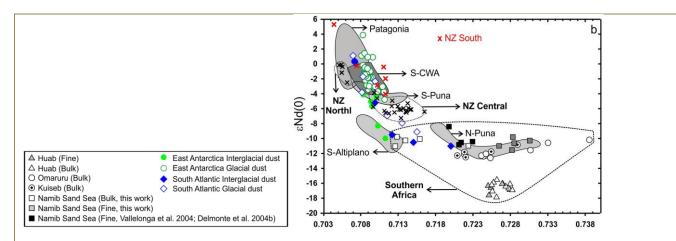


Fig. 5: (a) Comparison of Sr-Nd-Pb isotopic composition representing PSAs (SAF and SSA and NZ – New Zeeland) and glacial/interglacial dust from Epica Dome C (EDC). Data of East Antarctica dust from the interglacial (Holocene and MIS 5.5) and of glacial (MIS2), and data of sediment cores from the Atlantic sector of the Southern Ocean are also reported.

Task 2.4: Interpretation of the results for inorganic particle composition (ULB)

The SEM-EDS analyses on single particle collected on the Sigma 2 filters all along the transect provide a better understanding of the size and type of particle reaching Antarctica, especially the Antarctic coast and better constrain their origin.

As shown above within Figs. 4 and 5, the new REE and isotopic data set for snow samples collected along the transect, are very interesting. Southern Africa appears as another major dust source contributor for Antarctica, besides Southern South America, at least during modern periods. Further analyses comparing the interglacial/glacial signatures of the different ice cores available in the literature, will help to have a clearer picture about the intensity of dust production from the desert regions (Huab, Omaruru, Kuiseb and Namibia Sea Sand) of Southern Africa (Namibia) during past climatic periods. But our original isotopic results already suggest the SAF dust source was overlooked for the atmospheric depositions in EDC Antarctic ice cores and have stamped its isotopic footprint in southern hemispheric marine and ice cores during the Holocene.

Tracing dust remains challenging when long-range dust transports are involved around the Antarctic continent. Mixing of airborne particles from various regions often limits the use of geochemical tracers. Our novel statistical analysis based on comparisons between the REE patterns of Antarctica dust samples and those from all related PSAs allows discriminating and quantifying their potential respective contributions.

Task 3.1: Calculation of air mass trajectories (RMI)

The FLEXTRA model has been successfully applied to calculate air mass trajectories and a kmeans cluster analysis has been done based on several parameters. When the clustering is performed relying on the normalised latitude, longitude and altitude, four clusters of air mass origin were found. The cluster analysis has been done for the whole period and also for each season separately.

An example is shown in Figure 6 below. It shows the four air mass origin clusters (calculated over the whole period 2010-2019) for the austral summer season (December-January-February). All 10 days of the back trajectory calculation have been used for the clustering and all 10 days are included in the graphs.

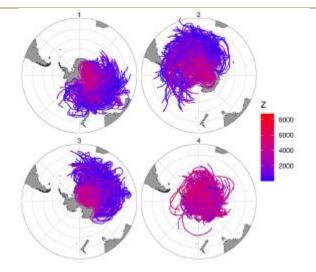


Figure 6: clusters of air mass origin, for austral summer, for 10 years of back trajectories starting at PEA station; Z is height in m asl

Some distinct features can be seen in the air mass origin clustering. Source regions from South America, Southern Africa and Australia were very limited. The Southern Ocean was a main source region, as was the Antarctic continent itself. For one of the clusters, the source region is mostly restricted to the region above the Antarctic continent. The average altitude along the trajectories in this cluster is higher compared to the average altitude of air coming from source regions over the Southern Ocean, indicating that this cluster corresponds to air subsiding from aloft. In Figure 7 below, a first analysis, showing the distribution of some parameters along the trajectories within the clusters is given.

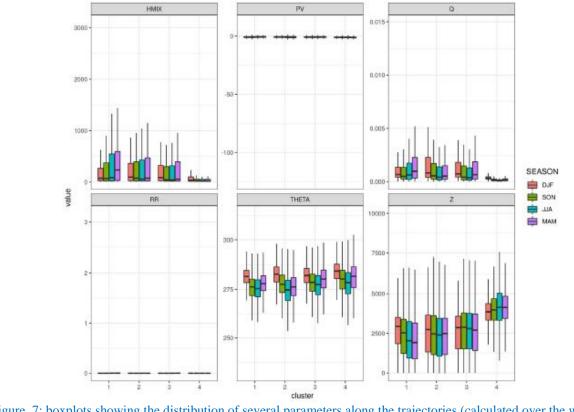


Figure 7: boxplots showing the distribution of several parameters along the trajectories (calculated over the whole period 2010-2019); Z = height above sea level; THETA = potential temperature; RR = total precipitation; Q = specific humidity; PV = potential vorticity; HMIX = mixing layer height

The higher potential temperatures during austral summers, the clear altitude feature of the air mass origin cluster 4 (with the Antarctic continent as contributor) and the low specific humidity during austral winter can be seen. However, the overall difference between the clusters and seasons is rather small. More detailed analyses are planned.

Task 4.1: Trace elements, micronutrients and atmospheric pollutants in Antarctica – their source regions, transport pathways, seasonal variations and relative importance of natural and anthropogenic compounds (RMI)

Air mass origin analyses have been done in order to investigate if the found source areas support the chemical fingerprint of rare-earth element data, found in certain snow samples, pointing to distinct source areas in Southern Africa or Southern South America, respectively. However, as mentioned in section 2, a clear discrimination between local and distal sources was not possible yet. However, with the most recent results from work package 3 and the results of the chemical analysis of organics, SVOCs, and inorganics, we will then be able to link particle properties to source regions, transport pathways and seasonal variations.

Task 4.2: Implications of found particle chemistry on cloud condensation and ice nuclei (RMI)

At PES, low INP concentrations (see Fig. 8) were obtained. Compared to the scarce literature data, the INP numbers for PES are at the lower limit. This shows the clear need to obtain more measurements, particularly as INP play an important role in ice formation in clouds and hence in precipitation formation. The new INP samples of season 2020/21 are therefore very important.

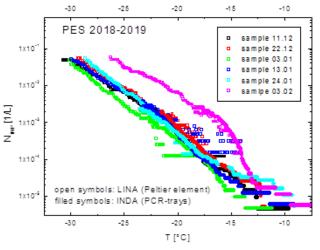


Figure 8: Number concentration of ice nucleating particles as function of freezing temperature

Task 5.1 : Network management (RMI) See section 2.

Task 5.2: Management of the chemistry database (RMI) See section 2.

Task 5.3: Publication of results to the scientific community, policy stakeholders and the general public (RMI, UGent, ULB, VUB)

See section 7 for an overview list of publications.

4. PRELIMINARY CONCLUSIONS AND RECOMMANDATIONS

Work package 1: Characterisation of the organic atmospheric composition (particulate matter and VOCs)

The obtained results are promising, and the developed methods prove to be robust and enable to compare data between the different sample years. Certainly, a unique dataset will be created containing a broad range of detected compounds and concentration levels.

Work package 2 Characterisation of the inorganic composition of atmospheric particles:

SEM-EDS analysis is a strong tool to characterize physically and chemically particles at the individual scale, especially in Antarctica were dust samples present a very low number of mineral particles and small grain-sizes. The present study highlights the very small particle size (< 5µm) of all samples, even near the Sør Rondane mountains, and a mineral profile suggesting a progressive change of dust provenance along the transect – with distal inputs at the coast, but also an influence from local rocks – underestimated in the current literature, even at the coast.

Complementary works need also to be performed to identify the anthropogenic inorganic compounds transported to Antarctica as atmospheric particles. In addition, a better appreciation of the Fe bioavailibility from its various forms/speciations could be performed (e.g., Shi et al., 2012; Visser et al., 2003) to better constrain dust Fe impact on the Austral Ocean and its implications on the ocean biological pump and the CO2 budget, on a global scale.

Within this study, we propose the first high-resolution reconstruction of dust provenance in modern snow from East Antarctica. This was made possible by the development of a new and innovative statistical approach applied to REE patterns of Antarctic dust and all the related dust PSA, which permits to trace and quantify dust origins across the Southern Hemisphere. Our results mark a breakthrough with respect to the usual vision of dust provenance in Antarctica. Our study shed new light on the role of SAF dust source areas that were, so far, overlooked. The challenging Sr, Nd and Pb isotopic analyses successfully confirm the SAF contributions to the dust deposits in East Antarctica, in addition to the SSA and local rocks. Our work allows untangling the mixing processes that leads identification of the PSA contributions for Antarctica remaining unclear without consensus inside the geochemist community.

Our study paves the way to reveal new key information from EDC and EDML ice cores during the LGIT period. Both ice core records, from separate locations in Antarctica, might indicate remarkably similar PSA mixing and transitions over time. Our studies are promising and shed new light on the evolution of the dust events and atmospheric circulation around Antarctica during the LGM and the Holocene, which surprisingly remain enigmatic today.

Work package 3 Air mass tracing by dispersion analysis of atmospheric transport:

The FLEXTRA trajectory model has been applied in order to investigate possible source regions and transport pathways into Antarctica of atmospheric particles and SVOCs. 10-days backward trajectories, starting from PEA, were calculated for a 10 year period. A k-means cluster analysis has been performed and four clusters of air mass origin were found. The cluster analysis has been done for the whole period and also for each season separately. Some distinct features can be seen in the air mass origin clustering. Within the constraints of this analysis, 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. There was one cluster of which the air mass originates almost completely from the Antarctic continent, corresponding to air subsiding from aloft.

Work package 4 Implications of the found results for atmospheric transport of trace elements, micronutrients and pollutants towards Antarctica and its closely associated Southern Ocean:

The role of changes in INP for clouds dominates over the role of changes in CCN (Solomon et al., 2018). Little is known about INP in the Antarctic region. Studies with contrasting results exist for the Southern Ocean. To the best of our knowledge, so far only one publication determined INP concentrations on Antarctica, at the South Pole (Ardon-Dryer et al., Atmos. Chem. Phys., 11(8), 4015-4024, doi:10.5194/acp-11-4015-2011), where concentrations of 1 per liter were found in the temperature range between -20°C and -24°C. At PES, we obtained clearly lower INP concentrations. These discrepancies show the clear need to obtain more measurements, particularly as INP play an important role in ice formation in clouds and hence in precipitation formation.

Work package 5 Coordination, database management and valorisation:

See section 2, achieved work.

General recommendations:

With respect to the analyses of chemical compounds, it can be said that the obtained results are promising and a unique dataset will be created containing a broad range of detected compounds and concentration levels. Our dataset will also lead to new insights on potential source areas. The prolongation of the CHASE project assures therefore the analysis of these very promising first results.

Based on SEM-EDS results, Sigma-2 passive sampling showed limitations in Antarctica encouraging active sampling even if this option implies very important logistical constraints. Active sampling will increase the quantity of the atmospheric particles enlarging the possibilities of geochemical tools to be applied to better specify their composition and origin. In the Sigma-2, the filter is affected by potential cycles of snow deposition/freezing event/melting event, which can lead to sea-salt accumulation on the filter and particle loss by molten snow streaming. An active sampling system will allow collecting more efficiently the dust on a shorter time period, when the meteorological conditions can be better monitored.

The role of changes in INP for clouds dominates over the role of changes in CCN. Little is known about INP in the Antarctic region and there is a clear need to obtain more measurements, particularly as INP play an important role in ice formation in clouds.

5. FUTURE PROSPECTS AND PLANNING

Overview of the foreseen activities and planning for next reporting year, taking into account the actual state of the work and the intermediary results

Work package 1: Characterisation of the organic atmospheric composition (particulate matter and VOCs) (UGent, VUB):

The remaining data of already performed analysis (QFF, 19-20 season sorbent tubes) will be analysed and compiled to obtain a complete dataset. The samples of the 20-21 need to be analysed with the developed methods and will also be added to this dataset.

The extraction and analysis of the passive PUFs can be continued as soon as the clean-up method is validated. Our aim is to submit a paper on the PAH topic encompassing both the active and passive sampling and a paper on the VOCs in Q2 and Q3 of 2021 respectively. Together with project partner RMI, it will be investigated if some of the observed trends in the dataset can be linked to air mass origin and air mass age. Further it will be investigated how observed differences between the passive sampling sites could be explained.

Work package 2: Characterisation of the inorganic composition of atmospheric particles (ULB) :

SEM-EDS analyses are planned to be performed on the filters collected from the passive Sigma-2 installations and an automatic active sampling (on PEA's southern science shelter). The mineralogical composition of East Antarctic dust should then be further detailed. By this approach of SEM-EDS single particle analysis, anthropogenic particle occurrence should be also confirmed and better identified. The plan is also to perform filtration, dissolution and analysis of trace element concentration of the remaining snow samples, especially the samples from 2020. Strontium, Nd and Pb isotopic ratios will be measured for the coast snow samples. Those results will significantly enlarge the geochemical and isotopic dataset of East Antarctica atmospheric deposition records. Finally, the innovative REE statistical model applied to our samples and the EDML ice core REE data should come out on outstanding publications, create new international collaborations and projects, and should pave the way for a better understanding of the air mass circulation nowadays and in the past in the Southern Hemisphere.

Work package 3: Air mass tracing by dispersion analysis of atmospheric transport: (RMI):

The FLEXTRA trajectory model will further be applied to identify air mass origin regions, including the cluster analysis. Especially, the properties along the trajectories within each cluster (e.g. meteorological parameters, altitude, time over ocean) will be further investigated, as well as particle properties between the clusters. For specific cases, also the FLEXPART dispersion model will be applied, e.g., to identify potential source regions for specific SVOC sample results.

Work package 4: Implications of the found results for atmospheric transport of trace elements, micronutrients and pollutants towards Antarctica and its closely associated Southern Ocean (RMI):

We will analyse the distribution of the measured atmospheric particle properties between and within the air mass origin clusters, derived in work package 3.. In addition, FLEXTRA and FLEXPART results will also be analysed for the air mass origin of specific events, e.g., when the aerosol total number concentration increased distinctly over short time, indicating new particle formation events. This can, e.g., also be applied to results of the VOC analyses. E.g., first analyses show distinct differences in the ratio between oxygenated and non-oxygenated polycyclic aromatic hydrocarbons for samples of different time periods.

The found physical and chemical characteristics of atmospheric particles will be combined with the results of the INP analyses, in order to better characterise the properties of ice nucleating and cloud droplet nucleating particles.

Work package 5: Coordination, database management and valorisation (RMI, UGent, ULB, VUB):

- The next Belgian Research Expedition to the PEA (November 2021 February 2022) will be planned similar to season 2020/21. However, the personnel on site (1 or 2 Pax) will be covered by the Brain2.0 project CLIMB, of which Alexander Mangold is coordinator and UGent partner. This work will mainly consist of recovering of the last passive samples, some snow samples and to recover and ship back all material.
- The CHASE website has been created and will be further developed
- Papers on results will be published in Geostandards and Geoanalytical Research and will be submitted soon to Nature: earth & environment, and to Science;
- Results of CHASE will be presented at vEGU 2021 (online meeting, presentations by Karen De Causmaecker), at the European Aerosol Conference (submitted contribution by Alexander Mangold) and other virtual conferences
- Further outreach activities like lectures at university, public talks, and blogs will be continued e.g., by Preben Van Overmeiren on 5 May 2021 on the UGent Faculty Research Days and on 18 May 2021 at an event of the UGent Alumni organization;
- The final scientific workshop will be organized, probably as an online event.

6. FOLLOW-UP COMMITTEE

Dates of the meetings and overview of the concrete contributions of the follow-up committee

Nadine Mattielli is keeping contact with Profs. Karine Deboudt and Pascal Flament (Laboratory of Physics and Chemistry of the Atmosphere (LPCA), Université du Littoral – Côte d'Opale, Dunkerque, France). They provide their expertise in aerosol characterisation by applying single-particle analysis (SEM-EDX) on the suspended atmospheric particles collected directly on filters (Sigma-2 and active pump samplers) and dust deposits (snow samples).

Nadine Mattielli is in email contact with <u>Dr. Volker Dietze</u> (German Meteorological Service, Research Centre Human Biometeorology, Air Quality Department, Freiburg, Germany) who provided the passive sampler equipment. They discussed the installation and improvement of the samplers.

A collaboration with <u>Paola Formenti</u>, Senior Scientist of the National Center for Scientific Research (CNRS) at LISA (France), opened new perspectives on a better understanding of the dust genesis from the main dust precursors (soils or loess from Southern South America or South Africa). Her work focuses on the optical and hygroscopic properties of aerosols and mineral dust in particular. She is head of the department where experiments on the CESAM simulation chamber are developed.

Nadine Mattielli collaborates with Prof. <u>James King</u> from Université de Montréal. He shared samples from his previous campaigns in Namibia, and A. Vanderstraeten collaborated with his PhD. Student Amélie Chaput at the CESAM lab for aerosol experiment from the Namibia sediment samples.

Nadine Mattielli closely collaborates with Prof. <u>Steeve Bonneville</u>, and G. Laruelle (from B-GeoSys, ULB) for the development of the statistical tools and the code required for the REE model set up. To have access to the REE data of prestigious Antarctic ice cores, such as EDML and EDC, N. Mattielli has contacted <u>P. Gabrielli</u> (Ohio state University) and <u>A. Bory</u> (from Université de Lille), who positively replied for a fruitful collaboration.

Nadine Mattielli, Christophe Walgraeve, Preben Van Overmeiren and Alexander Mangold are in contact with <u>Prof Annick Wilmotte</u> (University of Liège, Belgium). Her group and collaborators (e.g. of UGent, Brain-Be Microbian project) are studying the microbial diversity on deglaciated rocks, nunataks, or ridges in Antarctica. They are interested in how such taxa are distributed in Antarctica, e.g. via air transport. Our filter material might therefore be useful for microorganisms analyis. In addition, she is member of the Belgian delegation to the Committee for Environmental Protection to the Antarctic Treaty. Annick Wilmotte participated in the annual meeting of CHASE and ideas how to give parts of our samples to her have been exchanged.

Alexander Mangold is in email contact with <u>Prof. Nicole Van Lipzig</u> (KU Leuven). Within the Brain-Be Aerocloud project they collaborated on investigating the relationship between clouds, precipitation and aerosols in Antarctica. This collaboration is continued within the CLIMB BrainBe project. Within her group, the COSMO-CLM2 regional climate model has been adapted to simulate also the influence of different types of particles on the formation of clouds and precipitation.

Alexander Mangold is in email contact with <u>Dr. Heike Wex</u> (Leibniz Institute for tropospheric research, TROPOS, Leipzig, Germany) who is doing research on cloud formation, cloud processes and the aerosol particles involved in it. Her group is interested in the chemistry of the particles sampled within CHASE. A specific sampling setup has been sent to PE station during 2020/21 and several filters dedicated to analyses on the ice nucleating capabilities of the particles have been

collected. The new samples will be sent for analysis to Leipzig once arrived from Antarctica.

Thanks to <u>Stefania Gili</u>, the collaboration with <u>Prof. Diego Gaiero</u> (National University of Cordoba, Argentina) was effective through common publication (submitted soon at Nature: earth & environment). His research – dedicated to the dust characterisation and genesis in Chili and Argentine, is complementary to the CHASE objectives and the exchange of expertise and results will be beneficial for the outcome of the project. In September 2020, <u>Stefania Gili</u> was hired as senior scientist at Princeton university. She is in charge of the analytical lab and ice core projects. The close collaboration between Nadine Mattielli and Stefania Gili will be pursued and extended.

Nadine Mattielli and Alexander Mangold are in contact with <u>Prof François Fripiat</u> of ULB (Glaciology department). He is interested in the atmospheric chemistry of reactive nitrogen in Antarctica. The snow and especially the snow/ice core samples of CHASE are of interest to him – not only to potentially use them for his analyses but also to compare results of reactive nitrogen with the other chemistry results.

7. VALORISATION ACTIVITIES

7.1 PUBLICATIONS

Publications in peer-reviewed scientific journals:

Accepted or published:

- Aun, M., Lakkala, K., Sanchez, R., Asmi, E., Nollas, F., Meinander, O., Sogacheva, L., De Bock, V., Arola, A., de Leeuw, G., Aaltonen, V., Bolsée, D., Cizkova, K., Mangold, A., Metelka, L., Jakobson, E., Svendby, T., Gillotay, D., and Van Opstal, B.: Solar UV radiation measurements in Marambio, Antarctica, during years 2017–2019, Atmos. Chem. Phys., 20, 6037–6054, https://doi.org/10.5194/acp-20-6037-2020, 2020.
- Held, A. and A. Mangold, 2021, Measurement of fundamental aerosol physical properties, in: Th. Foken (ed.), Springer Handbook of Atmospheric Measurements, Springer, proofread accepted, in press.
- McCutcheon J., Lutz S., Williamson C., Cook J., Tedstone A., Vanderstraeten A., Wilson S., Stockdale A., Bonneville S., Anesio A., Yallop M., McQuaid J., Tranter M., Benning L. (2021) Mineral phosphorus drives glacier algal blooms on the Greenland Ice Sheet. Nature Communications 12, 570
- Vanderstraeten, A., Bonneville, S., Gili, S., Jong, J., Debouge, W., Claeys, P., Mattielli, N. (2020) First Multi-Isotopic (Pb-Nd-Sr-Zn-Cu-Fe) Characterisation of Dust Reference Materials (ATD and BCR-723): A Multi-Column Chromatographic Method Optimised to Trace Mineral and Anthropogenic Dust Sources. Geostandards and Geoanalytical Research, doi.101111ggr.12320.

In preparation:

- Gili T., Vanderstraeten A., Chaput A., King J., Gaiero D.M., Delmonte B., Vallelonga P., Formenti P., Di Biagio, C., Cazanau M., Pangui E., Doussin JF and Mattielli N., Importance of Southern Africa as source area for East Antarctica atmospheric deposition records during the interglacial periods. To be submitted for Nature: communications Earth & Environment (in June July 2021)
- Vanderstraeten A., Laruelle G., Gili T., Mattielli N., Bory A., Gabrielli P., Bonneville S. First highresolution reconstruction and quantification of dust provenance in East Antarctica over the last glacial-interglacial transition. To be submitted for Science Advances (in July 2021)

7.2 PARTICIPATION/ORGANISATION OF SEMINARS (NATIONAL/INTERNATIONAL)

Oral presentation, poster... and/or organisation of workshops, symposia etc.

Oral presentations:

- K. De Causmaecker, A. Mangold, C. Walgraeve, P. Van Overmeiren, N. Mattielli, S. Gili, and A. W. Delcloo, Identifying source regions at the Princes Elisabeth station in Antarctica, using dispersion modelling tools: a case study, European Geosciences Union General Assembly 2020 (online video conference), Abstract EGU2020-9127, 4-8 May 2020, Vienna, Austria, 2020.
- S. Gili, A. Vanderstraeten, M. Cazaunau, A. Chaput, J.-F. Doussin, C. Di Biagio, P. Formenti, J. S. King, A. Mangold, N. Mattielli, E. Pangui, P. Van Overmeiren and C. Walgraeve, The role of Southern Africa as a dust precursor to East Antarctica, European Geosciences Union General Assembly 2020 (online video conference), Abstract EGU2020-18441, 4-8 May 2020, Vienna, Austria, 2020.
- A. Vanderstraeten, N. Mattielli, G. G Laruelle, A. Bory, S. Gili, P. Gabrielli, S. Boxho and S. Bonneville. High-resolution statistical quantification of aeolian dust provenance in East Antarctica over the Last Glacial-Interglacial Transition. Abstract Goldschmidt Conference, 4-9 July 2021, Lyon, France.

Poster presentations:

- A. Mangold, 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.
- P. Van Overmeiren, S. Gili, A. Vanderstraeten, N. Mattielli, A. Delcloo, K. De Causmaecker, A. Mangold, K. Demeestere, H. Van Langehove, C. Walgraeve, Obtaining insight in atmospheric trace organic compound concentrations and trends in Dronning Maud Land, East Antarctica by means of long term passive and active air sampling, SCAR Open Science Conference (only online), Hobart, Australia, 3-7 August 2020.

7.3 SUPPORT TO DECISION MAKING (IF APPLICABLE)

The connection between scientific research on Antarctica and policy is largely managed by the Scientific Committee on Antarctic Research (SCAR). Belgium is a Full Member of SCAR, represented by the Belgian National Committee on Antarctic Research (BNCAR, <u>http://www.bncar.be/bncar/</u>). One meeting of BNCAR has been on 18 December 2020. Prof. Philippe Claeys and Dr. Alexander Mangold are members of BNCAR and have been following the meetings to ensure that all scientists involved are aware of the ongoing research. This is further strengthened via discussions with members of the follow up committee. In addition, CHASE scientists have joined the SCAR Action Group 'Input pathways of persistent organic pollutants to Antarctica, ImPACT (https://www.scar.org/science/impact/home/). On 31 March 2020 there has been a virtual meeting of the members of the SCAR ImPACT group.

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;

Blog on RMI's activities at Princess Elisabeth station: belatmos.blogspot.be;

Blog by Preben Van Overmeiren on the research activities during Belare 2020/21 (<u>https://ozone.meteo.be/projects/chase/belare2020-2021-campaign</u>)

Zoom-In (News-ticker of RMI) at the beginning of the season at PEA: <u>https://www.meteo.be/fr/infos/actualite/des-scientifiques-belges-suivent-de-pres-le-trou-record-dans-la-couche-dozone-en-antarctique</u>

TV documentary of the field season 2019/20 at PEA, including interviews with Preben Van CHASE passages Overmeiren and Stefania Gili of and on their field work: https://www.siouxproductions.be/les-gardiens-de-lantarctigue distributed on RTBF (December 2020 and three months available via streaming: https://www.rtbf.be/tipik/article/detail_les-gardiens-de-l-antarctiquevotre-nouvelle-serie-documentaire-debute-ce-lundi-sur-tipik?id=10634510) and VRT (March 2021);

Interviews with CHASE partners on PEA station website or on website of International Polar Foundation :

<u>http://www.antarcticstation.org/news_press/news_detail/preben_van_overmeiren_explains_aerosols_and_cloud_formation_in_antarctica_(</u>9 February 2021); short version also on UGent faculty website:

https://www.ugent.be/bw/en/news-events/news/expedition-antarctica-atmospheric-particles-preben-vanovermeiren.htm

http://www.antarcticstation.org/multimedia/picture gallery/traverse for the chase and climb projects (9 February 2021);

<u>http://www.polarfoundation.org/news_press/news/stefania_gili_on_studying_atmospheric_particles_in_antarctica</u> (1 September 2020);

http://www.polarfoundation.org/news press/news/alexander mangold contributions of research to polar science yopp jpcc (22 July 2020)

Video call of Preben Van Overmeiren during his stay at PEA, and 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

Video calls from PEA station (science liaison officer Henri Robert) with the BA GEOG and GEOL students, and with MA GEOG and GEOL in December 2020, and with MA ENVI for the course ENVIF529 on Feb 9., 2021

8. ENCOUNTERED PROBLEMS AND SOLUTIONS

Encountered problems/obstacles, adopted and/or envisaged solutions, unsolved problems

Due to the pandemic and its consequences in terms of delay for the sample delivery and restrictions of lab accessibility, the trace element and isotope analyses of dust samples from the last season 2019 fell behind schedule. In addition, Stefania Gili from Laboratoire G-Time was hired at Princeton University as senior scientist in September 2020, which is very good news but limits the data analysis. Nevertheless, following the PhD thesis of A. Vanderstraeten ended in September 2020, Sibylle Boxho started a new PhD thesis program as teaching assistant at ULB and made

progress in the filtration and analyses of the 2019 snow samples, at least for the trace element data. She is now busy with the sample preparations for the isotopic measurements.

Despite the Covid-19 pandemic, the Belare 2020/21 expedition to PEA station could take place. It meant however, a long quarantine period in Cape Town (RSA) for the expedition team, which prolonged from planned two weeks to four weeks due to meteorological conditions and needed quarantine time for other flight passengers or pilots. This shortened the planned time at PEA. In order to be able to do all foreseen field work, Preben Van Overmeiren stayed almost four weeks longer at PEA.

One pump for the active sampling for filter samples for inorganic analyses broke end of December and also the second one showed issues to generate a correct flow for the sampling. This sampling had therefore to be stopped end of December 2020 and only 3-4 samples could be collected. Nevertheless, it has been the fourth season during which samples could be collected. The pumps were shipped back to Belgium and will be repaired.

Due to the Covid-19 crisis, participation at (inter-)national scientific conferences is compromised. E.g., the International Conference on Aerolian Research has been postponed from 2020 to 2022. Other conferences took place virtually. Some with shortened agenda (e.g., SCAR Open Science Conference 2020), others kept the whole programme (e.g., EAC 2020). CHASE partners participated however in several virtual conferences, see section 7.

The prolongation of the CHASE project for one year until 15/04/2022 has been asked at BELSPO and has been agreed. This has been necessary in order to analyse all samples, including those from season 2020/21. Also, the samples form season 2019/20 arrived only in June 2020 in Belgium, later than usual, implying later than planned laboratory analyses. In addition, it allows MSc Preben Van Overmeiren to finish his PhD within the foreseen time of 48 months for his contract at Ghent University.

9. MODIFICATIONS COMPARED TO THE PREVIOUS REPORT (IF APPLICABLE)

9.1 PERSONNEL

KMI hired a new staff member: Karen De Causmaecker

Partner	Name	Nationality	Gender	Date of birth	Certificate	Year of graduation	Statute	Time implication in the project financed by BELSPO (in FTE)	Type of labour contract	Annual gross salary	Time implication in the project financed by other source(s) (in FTE)	Name(s) of the other funding source(s)	Remarks

9.2 COMPOSITION OF THE FOLLOW-UP COMMITTEE

See section 6.

10. REMARKS AND SUGGESTIONS

Concerning for example: the coordination, the use or valorisation of the results, personnel change ...

KMI:

Dr. Karen De Causmaecker worked on CHASE until 30 November 2020. Since December 2020 she is working on the closely related CLIMB (https://ozone.meteo.be/projects/climb) project. She is still pursuing CHASE-related tasks.

<u>ULB</u>:

Dr. Stefania Gili left ULB in 2020 for a new position at Princeton University, USA (since September 2020 there). She is still available for interpretation of samples and results. At ULB, Sibylle Boxho took over laboratory tasks for the ULB CHASE samples (however, not paid on CHASE).