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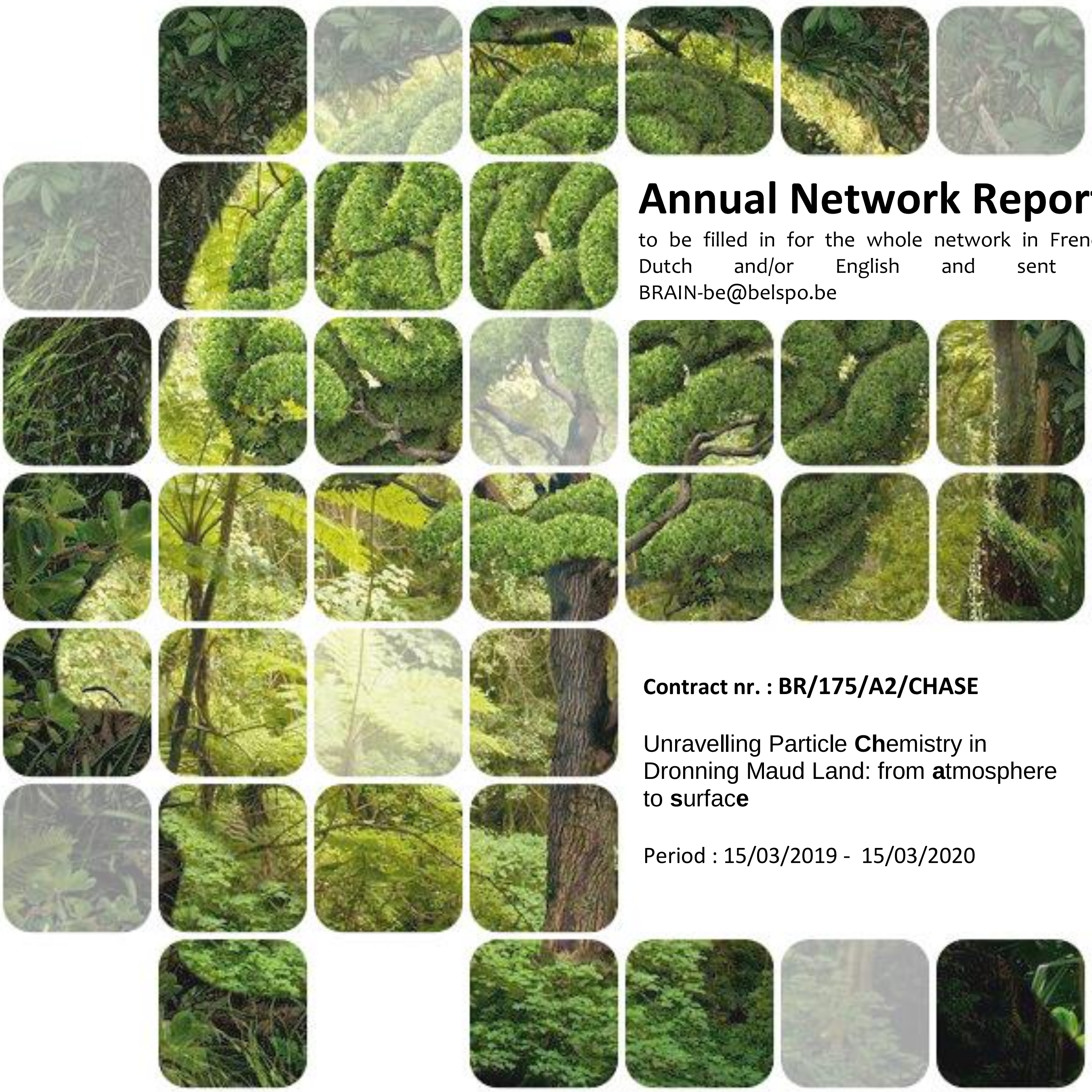

Annual Network Report

to be filled in for the whole network in French, Dutch and/or English and sent to BRAIN-be@belspo.be

Contract nr. : BR/175/A2/CHASE

Unravelling Particle **Ch**emistry in Dronning Maud Land: from **a**tmosphere to **s**urfac**e**

Period : 15/03/2019 - 15/03/2020

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NETWORK

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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 1st January 2017. In the table below, the submission date is counted from the 15th April 2017 (48 months until end of project in contract 15/04/2021.

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	PROG
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 2018/19 in February 2019, the samples arrived end of March 2019 at the partner's institutes and were stored in freezers and clean rooms for further analyses.

The samples for organic chemistry analysis of the period 2017 to November-December 2018 were analysed using GC-MS and PTR-MS. Before analysis on GC-MS a water purge technique was developed to remove excess water, as this damages the detector, without losing a significant amount of analytes. With GC-MS and a non-target screening method using AMDIS software about 70 compounds are consistently detected. Variance within one group of samples (i.e. from the same site) is low for most compounds while there are clear differences between sites. From the first results it is clear that PAH's and several VOC's were present in varying concentrations in the Antarctic atmosphere.

Thirteen snow samples plus 7 rock samples from the Sor Rondane mountains have been analysed in terms of Rare Earth Elements and Sr, Nd, Pb isotopes. Eight samples recovered from the Sigma-2 passive collectors and 6 filters from the Active sampling are "in-progress-stage". On the basis of our results, two papers are about to be submitted to peer-review journals. The new 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.

The FLEXPART model has been successfully applied to calculate air mass trajectories and for dispersion modelling. Potential source areas are possible to identify for distinct samples taken. Air mass origin analyses supported that the derived source areas from the chemical fingerprints of particles found in certain snow samples point to distinct source areas in Southern Africa or Southern South America, respectively.

The following preparations were undertaken for the 2019/20 field campaign at PE station:

- 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;
- Material for the sampling has been prepared and ordered. Cleaning and; preparation of the bottles and filters needed for sampling surface snow. UGent prepared an automated sampler for VOCs for autonomous winter operation.

Stefania Gili and Preben Van Overmeiren participated on the CHASE project in the BELARE 2019/2020 field campaign to Princess Elisabeth station and have been there from mid-November to just before Christmas 2019. They re-installed the pumps for active sampling near the station. In addition to the seven sites for passive sampling (see Figure 1) of atmospheric particles and volatile organic compounds installed last season, they succeeded in taking at two of the passive sampling sites (Frank Kenny North and Plateau) 3-m long ice cores for chemical analyses. At all seven sites, also surface snow samples were collected. The collected samples should arrive by end of April in Belgium and will be stored either at Ghent University or at Laboratoire G-Time (ULB) for further analyses. Stefania Gili and Preben Van Overmeiren helped in addition to supervise instruments of the former Brain-Be AEROCLOUD 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:

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

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

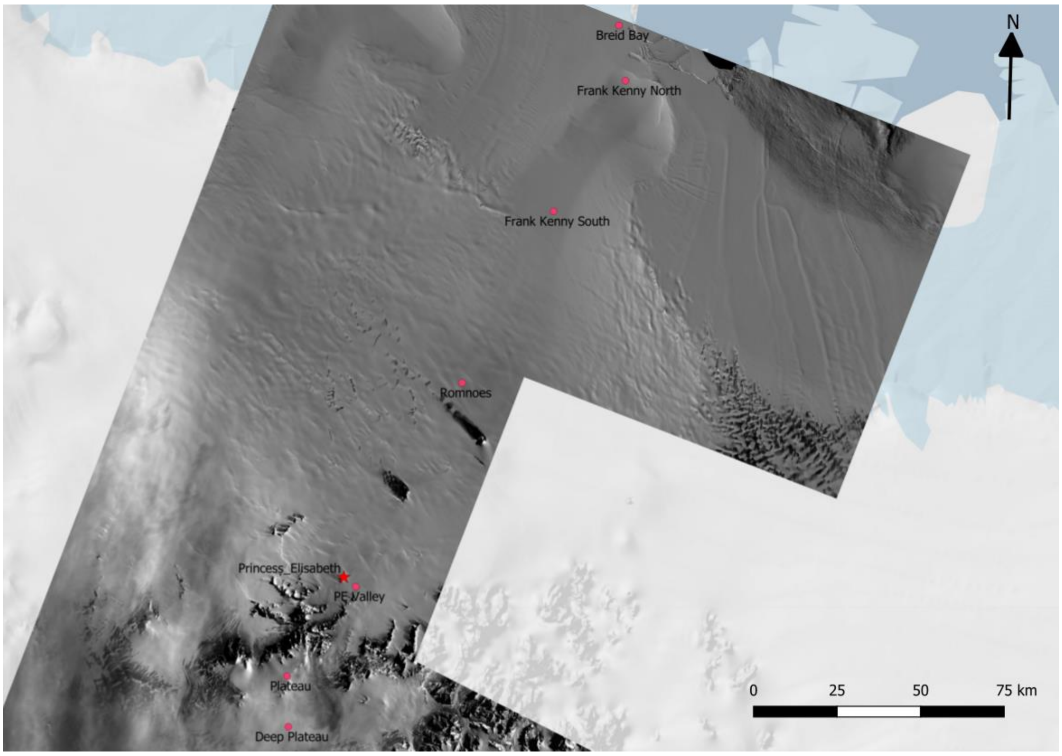

Figure 1: Location of the seven sampling sites where the CHASE passive samplers have been installed and where surface snow samples have been collected. 'Deep plateau', southward of PE, on the plateau; ii) 'Plateau', southward of PE, vicinity of the plateau; iii) 'PE valley', around 4 km eastward of PE; 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

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

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 2019-2020 austral summer the instrument ran for the 3rd 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. In addition, polymer sheet type passive samplers have been installed to sample organic micro-pollutants. Both 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 first sample set was acquired from the first 5 sites. During the 2019-2020 season all seven sites were visited for sample collection and exchange by Preben Van Overmeiren and Stefania Gili. New sorbent media were installed and are to be recovered during the next expedition. One site (Breid Bay) was removed because of too much snow accumulation.

Two 3-meter-long firn/ice cores where drilled at two sites (Frank Kenny North and Plateau) using a specialized titanium large diameter corer in order to be able to take a large enough volume of ice without contaminating the sample. The samples are part of a preliminary study for chemical analysis.

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 seven locations as the passive samplers for semi-volatile organic analysis mentioned before. In November, December 2019 the samples were collected by Preben and Stefania and new ones were installed. These samplers will collect VOCs over another year until recovery in November, December 2020.

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.

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

To characterize and quantify organic components sampled using the active (HVS) and passive methodology, an analytical approach using accelerated solvent extraction (ASE) was developed and validated. ASE has the advantage of reducing the required amount of solvent while obtaining high extraction efficiencies. The same method has been validated for the analyses of the quartz fibre filters which capture particulate matter. 80% of PUF samples from 2 seasons of high-volume sampling have been analysed with the developed method. Furthermore, a chromatographic method was developed to analyse oxy-PAHs, extending the range of detectable chemicals. The first set of axial passive samplers for VOC’s from the first campaign was completely analysed on two instruments. A method to purge excessive water from the tubes proved necessary before analysis on TD-GC-MS. The GC-MS was used to quantify and identify volatile compounds captured on the Tenax tubes. Compounds difficult to detect on GC-MS due to their low accumulated mass or specific structure are detected on the second instrument, PTR-MS.

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

A test sample originating from particles in surface snow samples near the coast was measured for $\delta^{13}C$ and $\delta^{15}N$. The sample was mostly composed of clay, with very limited organic material. The obtained $\delta^{13}C$ was -33‰. No reliable value was determined for $\delta^{15}N$. This value resembles those measured over urbanized area, but it is too early to draw any conclusions.

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

Because mass spectrometry creates datasets of tremendous size, statistical analysis is used to process the data and reduce dimensionality and revealing trends. Atmospheric processes and mechanism from literature are considered when analysing data.

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 μm pore-size Teflon filters has been done from December 2019 (CHASE team) through February 2020 (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, a total of 10 filter samples were recovered. These filter samples are already in Belgium and awaiting laboratory analysis.

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

During the season 2019-2020, the seven 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 site for the collection of inorganic particles over another year until the next campaign.

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 2019 by Stefania Gili at seven 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) near Romnoes, northward of PE; v) in between Romnoes and the coast; vi) 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 52 bottles of 10 L, i.e. a total of ~450 L of surface snow, have been collected. The bottles have been shipped back and are expected to arrive in Belgium at the end of April, 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.

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

Thirteen snow samples plus 7 rock samples from the Sor Rondane mountains have been analysed in terms of Rare Earth Elements and Sr, Nd, Pb isotopes. Eight samples recovered from the Sigma-2 passive collectors and 6 filters from the Active sampling are "in-progress-stage" and their chemical/isotopic compositions are expected to be measured in May/June 2020. On the basis of our results, two papers are about to be submitted to peer-review journals (see section 7). The analyses of the first snow samples allowed us to know with more accuracy the volume of snow required for proper sampling at each site, since the deposition and accumulation rates vary along the ~230 Km transect.

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

Task 3.1: Calculation of air mass trajectories (RMI)

The FLEXPART model has been applied to calculate air mass trajectories and also for dispersion modelling of air masses. These calculations have been done mainly for two snow samples, collected in December 2014 and January 2016 near the coast, in Dronning Maud Land, East Antarctica. Further, the model has been used to investigate if air masses of the Australian bush fires of 2019/2020 could reach this part of Antarctica.

<u>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)</u>

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 which points to distinct source areas in Southern Africa or Southern South America, respectively. The respective results will be used for a publication in Geochemical Perspective Letters.

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; https://www.atmos-chem-phys.net/19/275/2019/).

Based on the first INP test samples of season 2017/18, the sample time for INP filter collection was prolonged to 10 days for the campaign 2018/19. A total of 6 samples could be collected. During spring 2019 these samples have been analysed at TROPOS, Germany for the number of ice nucleating particles.

<u>Task 5 Coordination, database management and valorisation (RMI, UGent, ULB, VUB)</u>

Task 5.1: Network management (RMI)

Project coordination is led by the Royal Meteorological Institute. A general meeting of all CHASE partners took place on 24 March 2020 via an online meeting (due to Covid-19 travel and gathering restrictions). Several additional meetings of partners took place before the BELARE 2019/20 campaign and after it, in order to prepare the campaign and to give feedback on the campaign.

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 chase.meteo.be

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, and to Geostandards and Geoanalytical Research. Manuscripts are planned to be submitted to Earth and Planetary Science Letters and to Geochemical Perspective Letters.
Partners of CHASE presented results on several international scientific conferences. The general public has been addressed at several occasions. E.g., Nadine Mattielli presented CHASE at a TV show of a Brussels TV channel in February and March 2020. Blogs have been maintained during the field campaign by Stefania Gili and Nadine Mattielli www.bncar.be), Preben Van Overmeiren (https://www.ugent.be/bw/gct/en/research/envoc/blog) and by Alexander Mangold (belatmos.blogspot.be). For a detailed list of publications, 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)

No changes were made to the 6 passive sampling sites remaining. The previously developed method for analysing PAH's in high volume PUF's was validated for the analysis of particle associated PAH's on the quartz fibre filters. The first 2 sample years are partially analysed, as the work was interrupted due to the current COVID-19 situation.

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

No new additions were made to the passive sampling sites. For the 2019-2020 season an autonomous active sequential VOC sampler was developed by the EnVOC lab. This sampler is designed to run without user interaction for a whole year, including the Antarctic winter, and is currently installed for trial at the princess Elisabeth station. At the time of writing this report, it is still well operating.

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

The samples of the period 2017 to November-December 2018 were analysed using GC-MS and PTR-MS with the validated methods. Before analysis on GC-MS a water purge technique was developed to remove excess water, as this damages the detector, without losing a significant amount of analytes. With GC-MS and a non-target screening method using AMDIS software about 70 compounds are consistently detected. Variance within one group of samples (i.e. from the same site) is low for most compounds while there are clear differences between sites. This is visualised using PCA in Figure 2 below.

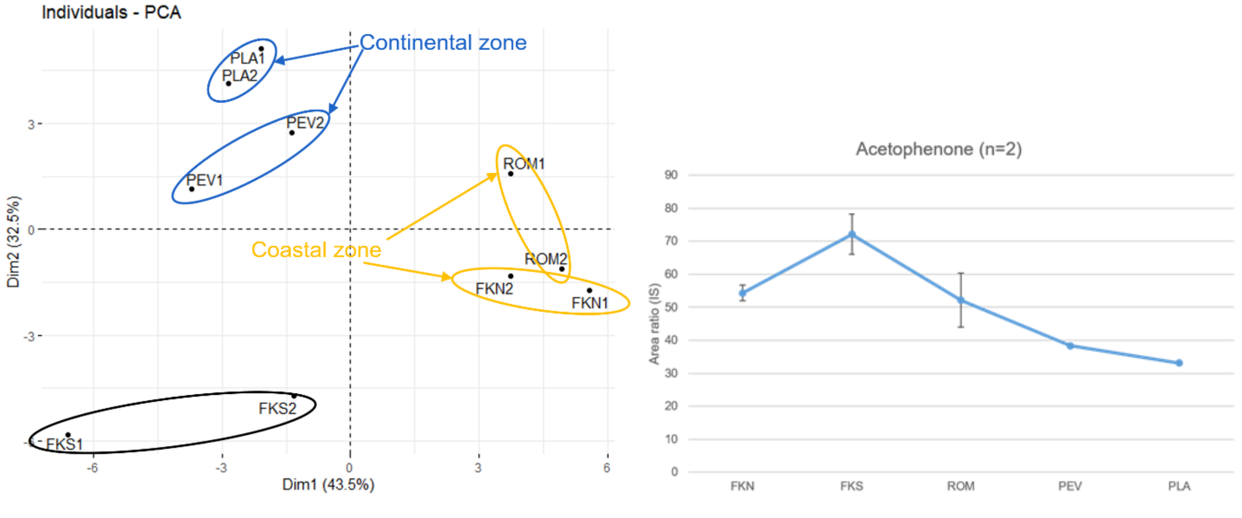

Figure 2: PCA analysis of the GC-MS data obtained from Tenax TA tubes exposed during one year from Nov/Dec 2017 to Nov/Dec 2018 on the left. Response of acetophenone (oxidation product of ethylbenzene) relative to the internal standard (toluene-D8) for different sample sites on the right.

Using the PTR-MS technique compounds can be detected which are too low in abundance or difficult to detect on TD-GC-MS due to their structure. Using an innovative coupling the sorbent tubes can be analysed on the PTR-MS instrument. In this analysis we found various small molecules such as methylglyoxal which are important breakdown products from atmospheric oxidation of aromatic pollutants. By combining the results of mass spectrometric analysis with literature study most compounds can be related with the oxidation of various primary pollutants. The presence of oxygenated species indicates it is very unlikely that samples are influenced by local emissions.

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

A test sample originating from particles in surface snow samples near the coast was measured for $\delta^{13}C$ and $\delta^{15}N$. The sample was mostly composed of clay, with very limited organic material. The obtained $\delta^{13}C$ was -33‰. No reliable value was determined for $\delta^{15}N$. This value resembles those measured over urbanized area, but it is too early to draw any conclusions.

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

Ion chromatographic purifications of Sr, Nd and Pb isotopes have been performed on 6 samples from the active pump sampling. Three out of 6 samples correspond to the austral summer 2017-2018 and the other 3 to the period 2018-2019. The main goal is to see any possible difference in the two-year sampling. Due to the Covid-19 situation, the analyses of these samples using our HR-MC-ICP-MS at the ULB have been delayed.

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

A lot of energy and time were spent to set up the methodology to dissolve the atmospheric particles from the filters, with attention to minimize the risk of particle loss and contamination risk. From the Sigma-2 passive collectors installed in the 7 different sampling sites during the CHASE campaigns (i.e. (1) 'PE valley', around 4 km eastward of PE, (2) 'Plateau', southward of PE, (3) Deep Plateau; (4) 'Romnoes'; (5) 'Frank Kenny South'; (6) 'Frank Kenny North', near the coast and (7) Breid Bay), 8 samples have been already processed through acid dissolution and are ready to be analysed for trace element concentrations (Rare Earth (REE) and Metal Trace Elements) with the HR-ICP-MS. These analyses will take place in the near future together with those of the active sampling samples mentioned above. These results are promising since we will be able to see if some of the discrepancies observed last year between the filter's results and the ones from the snow samples, are to be seen again or not. So far, our explanation to the encountered differences could be the successive repetitions of snow or ice accumulation and melting events inside the passive collectors throughout the year, which might imply moving of atmospheric particles out of the filters.

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

The 230 L (in volume) of snow collected by Nadine Mattielli during November-December 2017 and the 250 L collected by Stefania Gili during November-December 2018, were melted by Stefania Gili at the Laboratoire G-Time and processed for chemical and isotope analyses (Sr and Nd). Same as before, samples from the 7 sampling sites were analysed for trace element concentrations and radiogenic isotope compositions. The obtained results (e.g. Rare Earth elemental ratios and Sr, Nd and Pb isotopes; see Figs. 3 and 4 below) are part of two "in progress" manuscripts to be send in the next months to peer review journals (GPL and EPSL, respectively) by Stefania Gili and Aubry Vanderstraeten as the main authors. An additional paper dedicated to the analytical methodology specifically developed for isotopic analyses of dust samples and appropriated reference materials is now accepted and will be published in GGR journal. As a whole, our results help to better pinpoint the origins of dust reaching Antarctica and understand their temporal evolution. Specific methodologies were developed to work with very low-mass dust samples, such as it is the case in Antarctica; in addition, a novel statistical model of Rare Earth Element patterns is proposed. We have compared the REE signature for the Antarctic samples vs. the REE fingerprint of different potential source areas from the Southern Hemisphere and found that in this sector of East Antarctica (inland area) a local signature, from the Sor Rondane mountains prevails in combination with a distal signature from dust regions in Southern South America. In addition, for the first time a Southern Africa signature was identified for the coastal Antarctic sampling sites (Fig.3). These results are confirmed by the radiogenic isotopes (Fig. 4), where a similar isotopic signature in terms of Sr and Nd for both the snow modern dust samples of Antarctica and Southern Africa is observed.

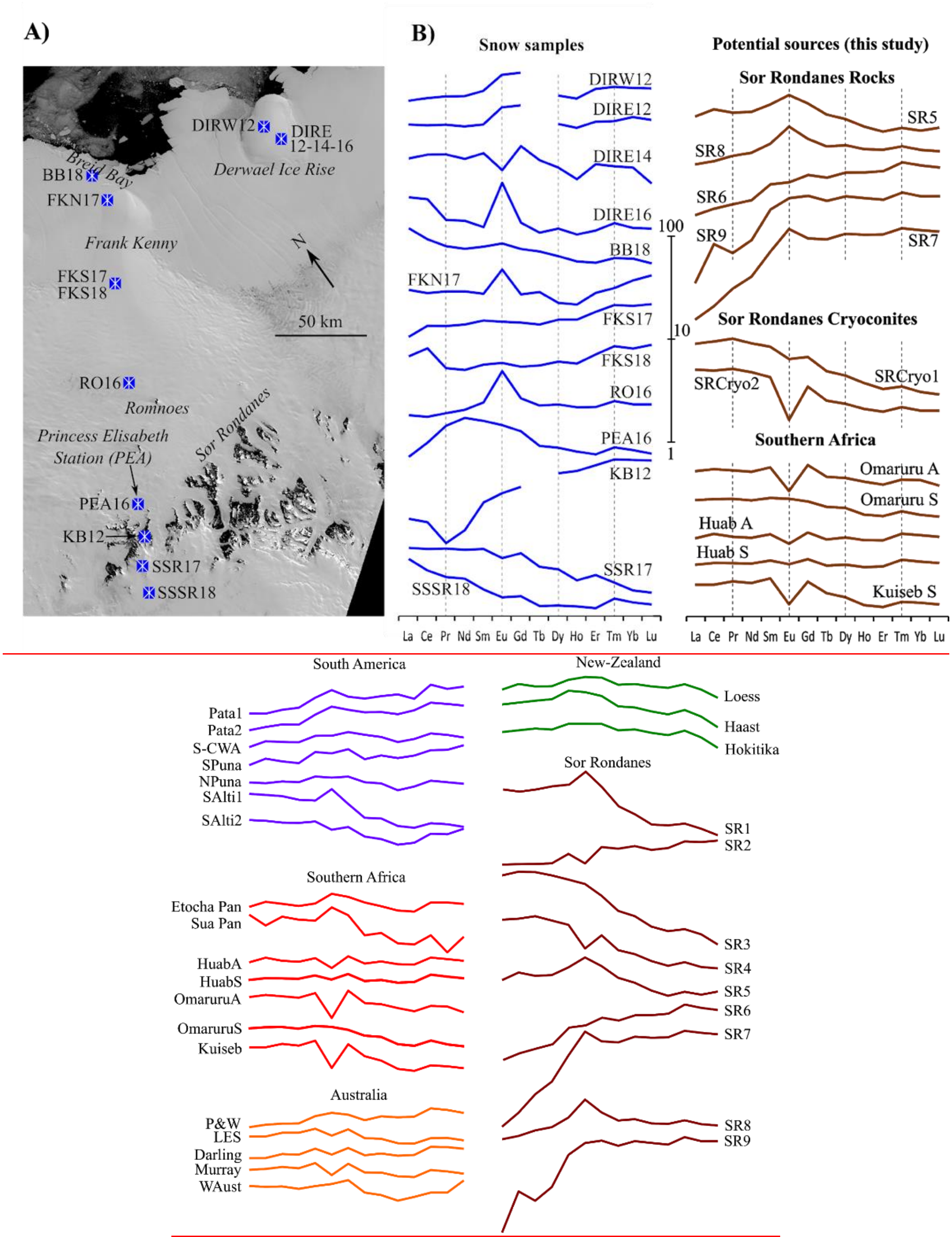

Fig.3: (A) ESA-Sentinel images (January-2020) indicating snow sampling sites along the transect. Number in the sample name corresponds to the year of sampling. (B) (on the left) REE patterns of all samples analysed in this study; (on the right and below) REE patterns of geological materials from dust precursor area (rocks, cryoconites, and re-suspended dust from sedimentary deposits) and REE profiles reported from literature.

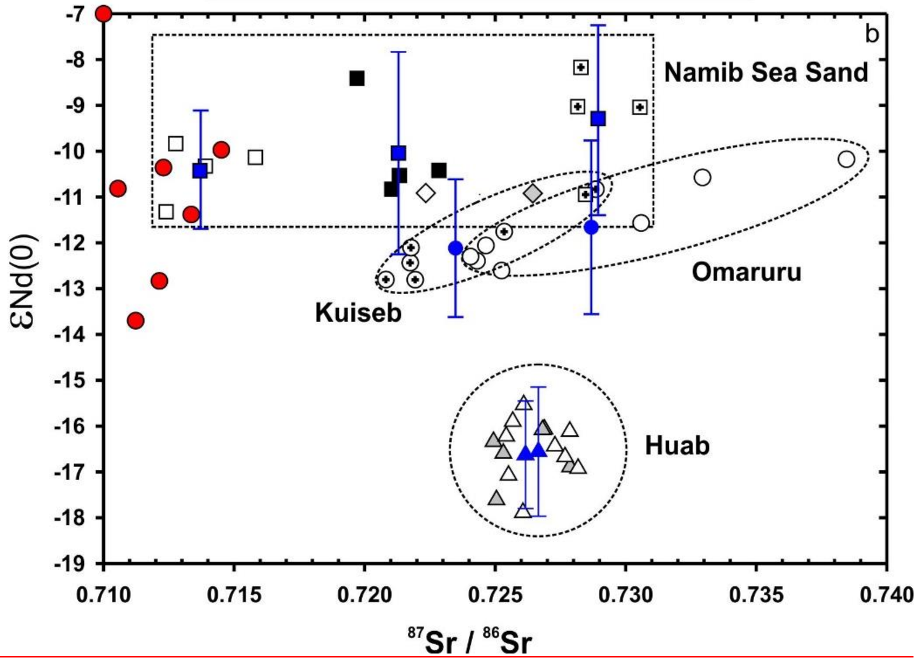

Fig. 4: Sr and Nd isotopic ratios for the modern dust collected in snow samples from our study transect in East Antarctica (red dots) and the signature of four dust source areas in Southern Africa (Huab, Omaruru, Kuiseb and Namibia Sea Sand). The blue lines represent the 2-sigma error bars for Nd composition.

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

As shown above within Figs. 3 and 4, the new 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.

Task 3.1: Calculation of air mass trajectories (RMI)

The FLEXPART model has been successfully applied to calculate air mass trajectories and for dispersion modelling. Further, the model has been used to investigate if air masses of the Australian bush fires of 2019/2020 could reach this part of Antarctica.

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)

For a paper, to be submitted to Geochemical Perspective Letters, air mass origin analyses have been done in order to investigate if the found source areas support the chemical fingerprints of particles found in certain snow samples which point to distinct source areas in Southern Africa or Southern South America, respectively. Figure 5 shows results of the atmospheric dispersion modeling for two collected snow samples: DIR14 (7 Dec 2014; left) and DIR16 (15 Jan 2016; right). The graphs show the percentage of particle mass originating from a certain region. It nicely can be seen that the majority of particle mass originates from regions nearby the Antarctic coast and that there was a circular-like pattern around Antarctica with decreasing contribution when moving to lower latitudes. For the DIR14 sample, the influence of cyclonic activity for transporting air masses (and also particles) can clearly be seen (the more reddish colours tracing the cyclonic form). On 5 and 6 December 2014, there was indeed a storm in the region where the sample has been taken. The air mass origin analysis supports that the fingerprint of rare earth elements of samples DIR14

(Southern Africa, Namibia) and DIR16 (Southern South America) are different. For the DIR14 sample the Namibian coast and parts of Southern Africa are clearly within the potential source area whereas this is distinctly less for the DIR16 sample. For the DIR16 sample, Southern South America and the Altiplano region is more distinctly a potential source area than for the DIR14 sample.

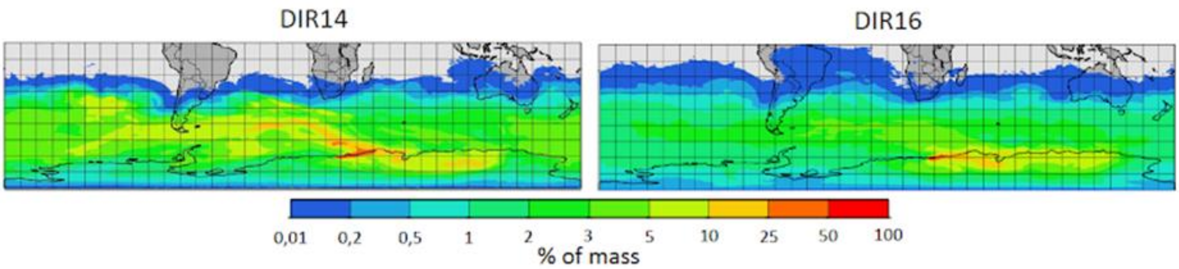

Figure 5: Results of the FLEXPART atmospheric dispersion modeling for two samples: DIR14 (left graphs) and DIR16 (right graphs). The upper graphs show the percentage of particle mass (of initialization specification) originating from a certain region

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

Based on the first INP test samples of season 2017/18, the sample time for INP filter collection was prolonged to 10 days during the season 2018/19. A total of 6 samples could be collected. During spring 2019 these samples have been analysed at TROPOS, Germany in order to obtain atmospheric concentrations of ice nucleating particles (INP). At PES, low INP concentrations (see Fig. 6) 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.

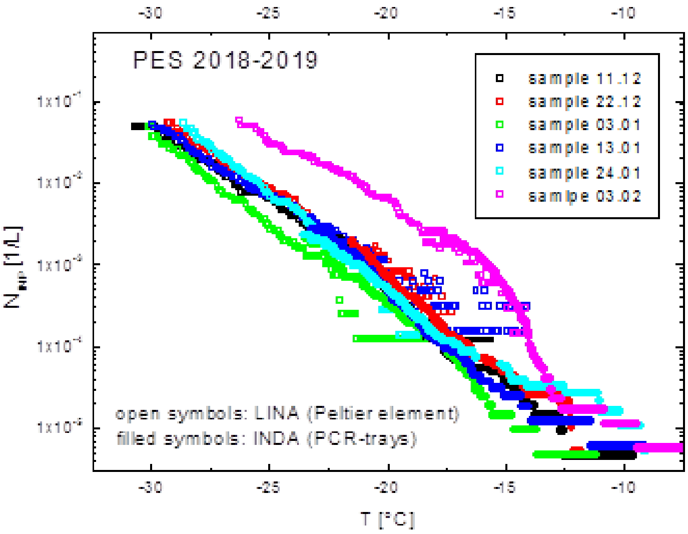

Figure 6: Number concentration of ice nucleating particles as function of freezing temperature

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)
From the first results it is clear PAH's and several VOC's were present in varying concentrations in the Antarctic atmosphere. Further work needs to be performed to try and find possible atmospheric breakdown products and pathways. This includes actively looking in the obtained data for key oxidized products known from the literature as well as pinpointing a mechanism for some currently unexplained chemicals. Afterwards the discovered trends can be tested with the obtained knowledge. Re-analysing samples for oxygenated-PAHs will be necessary to achieve more knowledge on the distribution of PAH's between gas phase and aerosol phase.

Work package 2 Characterisation of the inorganic composition of atmospheric particles:
Thirteen snow samples plus 7 rock samples from the Sor Rondane mountains have been analysed in terms of Rare Earth Elements and Sr, Nd, Pb isotopes. Eight samples recovered from the Sigma-2 passive collectors and 6 filters from the Active sampling are about to be analyses. On the basis of our results, two papers are almost finalized to be submitted to peer-review journals (see section 7). The analyses of the first snow samples allowed us to know with more accuracy the volume of snow required for proper sampling at each site, since the deposition and accumulation rates vary along the ~230 km transect.

As shown above within Figs. 3 and 4, the new 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.

Work package 3 Air mass tracing by dispersion analysis of atmospheric transport:
The FLEXPART model has been successfully applied to calculate air mass trajectories and for dispersion modelling. Potential source areas are thus possible to identify for distinct samples taken.

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:
Air mass origin analyses have been done in order to investigate if the found source areas support the chemical fingerprints of particles found in certain snow samples which point to distinct source areas in Southern Africa or Southern South America, respectively. Figure 5 shows results of the atmospheric dispersion modeling for two collected snow samples. The air mass origin analysis supports that the fingerprint of rare earth elements of samples DIR14 (Southern Africa, Namibia) and DIR16 (Southern South America) are different. For the DIR14 sample the Namibian coast and parts of Southern Africa are clearly within the potential source area whereas this is distinctly less for the DIR16 sample. For the DIR16 sample, Southern South America and the Altiplano region is

more distinctly a potential source area than for the DIR14 sample.

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, where Bigg (1973) obtained much larger INP concentrations than McCluskey (2018). To the best of our knowledge, so far only one publication determined INP concentrations on Antarctica, at the South Pole (Ardon-Dryer et al., 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 (see Fig. 6). These discrepancies in the scarce literature data 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

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.
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.
McCluskey, C. S., et al. (2018), Observations of Ice Nucleating Particles Over Southern Ocean Waters, Geophys. Res. Lett., 45(21), 11989-11997, doi:10.1029/2018gl079981.
Solomon, A., et al. (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.

Work package 5 Coordination, database management and valorisation:
See section 2, achieved work.

General recommendations:
The first results of Chase show that trace inorganic elements can be identified and also VOCs. The applied methods are therefore successful. The Chase partners gathered so far samples during three consecutive Antarctic campaigns and started a promising and unique time series of measurements of the Antarctic particle chemistry. Such measurements are encouraged by the Scientific Committee of Antarctic Research (SCAR). A specific Action Group has been established: Import pathways of Persistent Organic Pollutants to Antarctica (ImPACT; https://www.scar.org/science/impact/home/). One goal is establishing a long-term monitoring of chemistry of organic pollutants and its pathways. Our atmospheric measurements are very important. Several initiatives and projects exist, but most of them cover the marine compartment or biota and there are only few respective atmospheric measurements.

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):
Once the samples collected during the previous expedition (November-December 2019) will arrive in Ghent, the established methods can be applied to analyse these and complete the dataset. Further work will be done in data-analysis and working towards a scientific publication. Extra effort will be put in analysing the different PUFs samples remaining from previous campaigns. The autonomous sampling of VOC's is bound to give a unique insight in how VOC patterns evolve during the year. For an extension project the acquired knowledge will be used to build a 2^{nd} autonomous sequential VOC sampler to be deployed in a remote area in East-Antarctica starting

November-December 2020.
A new set of surface snow samples has to be obtained, and C and N isotopic value should be measured on raw samples from the 2019-2020 campaign.

Work package 2: Characterisation of the inorganic composition of atmospheric particles (ULB) :
Beginning of May 2020, the melting of the ~450 L (in volume) of snow and its filtration to recover the new atmospheric particles from the 2019/20 sampling campaign will begin. At the same time, the 8 samples from the passive collectors and the 6 from the active sampling will be measured by the HR-ICP-MS at the VUB for trace element analyses and on the Nu Plasma II MC-ICP-MS at ULB for isotopes.

Work package 3: Air mass tracing by dispersion analysis of atmospheric transport: (RMI):
The dispersion modelling and calculation of air mass trajectories will be continued. These analyses will include the samples taken and also clustering of potential source areas and atmospheric pathways.

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):
For the field campaign 2020/21 at PES there will be again INP filter samples collected. Further, the analyses of the dispersion modelling and of the back trajectory calculations will be combined with the found results of the chemical analyses of the (in-)organic samples.

Work package 5: Coordination, database management and valorisation (RMI, UGent, ULB, VUB):
- The next Belgian Research Expedition to the Princess Elisabeth station (November 2020 – February 2021) will be planned like it has been done for season 2019/20. However, the personnel on site (2 Pax) will be covered by the new Brain2.0 project CLIMB, of which Alexander Mangold is coordinator and UGent partner. This personnel will take care also of all CHASE related work.
- A CHASE website has been created and is hosted at RMI (chase.meteo.be). The website will be further developed in order to give information on samples and availability of data.
- Papers on results will be submitted to Geochemical Perspective Letters, and Earth and Planetery Science Letters.
- Results of CHASE should be presented at EGU 2020 (online meeting, accepted presentations for Stefania Gili and Karen De Causmaecker), ICAR 2020 (International Conference on Aeolian Research) and at the SCAR Open Science Conference 2020 (probably virtual meeting). However, due to the current worldwide Covid-19 situation, the attendance to these conferences seems to be compromised.
- Further outreach activities like public talks, and blogs will be continued. A video on the project consisting of movie clips shot by Preben Van Overmeiren during the 2018 and 2019 campaigns is in the course of realization.

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). Two SEM-EDX analytical sessions were organized at Dunkerque for the single particle characterizations and gave chemical composition and mineralogical characterization of the atmospheric particles.

Nadine Mattielli is in email contact with Dr. Volker Dietze (German Meteorological Service, Research Centre Human Biometeorology, Air Quality Department, Freiburg, Germany) who provided the passive sampler equipment. Volker Dieze also visited ULB in autumn 2018 and winter 2019. They discussed the installation and improvement of the samplers.

A collaboration with Paola Formenti, Senior Scientist of the National Center for Scientific Research (CNRS) at LISA (France), opens 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, Christophe Walgraeve, Preben Van Overmeiren and Alexander Mangold are in contact with Prof Annick Wilmotte (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 Annick have been exchanged.

The employment of Preben Van Overmeiren opened the perspective to collaborate with Prof. Laszlo Vincze UGENT), the former supervisor of Preben. Prof Vincze is doing research on (interstellar) dust characterisation using Synchrotron X-Ray analysis. There were some preliminary tests, however the results were not promising enough in order to further explore this possibility.

Alexander Mangold met before the Antarctic field campaign with Prof. Nicole Van Lipzig (KU Leuven). Within the Brain-Be Aerocloud project they are both collaborating on investigating the relationship between clouds, precipitation and aerosols in Antarctica. 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 Dr. Heike Wex (Leibniz Institute for tropospheric research, TROPOS, 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 2018/19 and several filters dedicated to analyses on the ice nucleating capabilities of the particles have been collected. The samples have been analysed and results are described further above.

The employment of Stefania Gili opened the perspective to collaborate with Prof. Diego Gaiero (National University of Cordoba, Argentina), the former supervisor of Stefania Gili. He is doing research on dust characterisation and genesis in Chili and Argentine, the areas foreseen as the main sources of dust deposits in Antarctica. His research is complementary to the CHASE objectives and the exchange of expertise and results will be beneficial for the outcome of the project.

Nadine Mattielli and Alexander Mangold are in contact with Prof François Fripiat of ULB (Glaciology department). He is interested in the atmospheric chemistry of reactive nitrogen in Antarctica. The snow and especially the snow/ice core samples of CHASE are of interest to him – not only to potentially use them for this 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:

- Aun, M., Lakkala, K., Sanchez, R., Asmi, E., Nollas, F., Meinander, O., Sogacheva, L., De Bock, V., Arola, A., de Leeuw, G., Aaltonen, V., Bolsee, D., Cizkova, K., Mangold, A., Metelka, L., Jakobson, E., Svendby, T., Gillotay, D., and Van Opstal, B.: UV radiation measurements in Marambio, Antarctica during years 2017–2019 in a wider temporal and spatial context, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-896, in review, 2019.
- Vandersraeten, A; S. Bonneville; S. Gili; W. Debouge; P. Claeys and N. Mattielli. 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:10.1111/ggr.12320.
- Gili et al., “in preparation” for submission to Earth and Planetary Science Letters, Reviewing the role of Southern Africa as a dust contributor to East Antarctica.
- Vanderstraeten et al., “in preparation” for submission to Geochemical Perspective Letters, Identifying and quantifying dust sources in northeast Antarctica using REE.

7.2 PARTICIPATION/ORGANISATION OF SEMINARS (NATIONAL/INTERNATIONAL)

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

Oral presentations:

- Mangold, A., Q. Laffineur, A. Delcloo, V. De Bock, C. Hermans, F. Hendrick, P. Herenz, H. Wex, A. Gossart, N. Souverijns, N. van Lipzig, I. Gorodetskaya, and H. De Backer, Atmospheric aerosol characterisation and relations to clouds and precipitation in Dronning Maud Land, East Antarctica, European Aerosol Conference 2019 Abstract O10_F1_A05, 25–30 August, Gothenburg, Sweden, 2019.
- Vanderstraeten, A., S. Gili, A. Mangold, S. Bonneville, V. Dietze, C. Walgraeve, P. Van Overmeiren, C. Berclaz, S. Goderis, N. Mattielli, Chasing dust in Dronning Maud Land, East Antarctica: A trace element perspective, 20th Congress of the Internatinonal Union for Quarternary Research (INQUA), Dublin, Ireland, 25-31 July, 2019.
- Gili, S. Chasing dust in Dronning Maud Land, East Antarctica: A geochemical perspective. *Seminar, Ghent University,* 10 May 2019
- Van Overmeiren P., Gili S., Mattielli N., Mangold A., Demeestere K., Van Langenhove H., Walgraeve, C., Organic compounds in the Antarctic atmosphere : first results of the 2017 and

2018 field sampling campaign, 'Antarctic climate symposium', Belgian Science Policy BRAIN-be Aerocloud event, Brussels, 10 may 2019.

Poster presentations:

- Souverijns, N., A. Gossart, A. Mangold, Q. Laffineur, P. Herenz, H. Wex, I.V. Gorodetskaya, G. Eirund, A. Possner and N.P.M. Van Lipzig, The impact of aerosols on clouds in the pristine environment of East Antarctica, European Geosciences Union General Assembly 2019, Geophysical Research Abstracts, Vol. 21, EGU2019-9838, 7-12 April 2019, Vienna, Austria, 2019.
- Mangold, A., Q. Laffineur, A. Delcloo, V. De Bock, C. Hermans, P. Herenz, H. Wex and H. De Backer, Atmospheric Aerosol Characterisation at Princess Elisabeth Station, Antarctic Climate Symposium, Belgian Science Policy BRAIN-be Aerocloud event, Brussels, 10 May 2019.
- Mangold, A., Q. Laffineur, R. Van Malderen, V. De Bock, C. Hermans, K. Nys, M. Verbruggen and H. De Backer, Total Ozone UV and radio sounding measurements at Utsteinen, Antarctic Climate Symposium, Belgian Science Policy BRAIN-be Aerocloud event, Brussels, 10 May 2019.

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, http://www.bncar.be/bncar/). 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/).

7.4 OTHER

- Mangold, A., Measurements of atmospheric particles in Antarctica: what even a low number of particles can tell us about the Antarctic atmosphere, Nanoparticles and air quality (talk in French), Seminar of TSI, JJBos, 21 May 2019, Ottignies-Louvain-La-Neuve, Belgium.
- Mangold, A., What have we learned from our atmospheric research at Princess Elisabeth station, Antarctica?, public lecture at RMI, 10 October 2019, Brussels, Belgium.
- Blog by Stefania Gili and Nadine Mattielli on research activities during Belare 2019/20: (www.bncar.be)
- Blog on RMI's activities at Princess Elisabeth station: belatmos.blogspot.be
- Blog by Preben Van Overmeiren on the research activities during Belare 2019/20 (https://www.ugent.be/bw/gct/en/research/envoc/blog)
- 2 videocalls from the PEA station to secondary schools by Preben Van Overmeiren
- Stefania Gili created a Twitter account and regularly posted from PEA the news from CHASE last mission.
- Stefania Gili and Preben Van Overmeiren were interviewed during the first week of the arrival at PEA for the local Belgium TV. Interview will be in air June/July 2020
- Prof. Nadine Mattielli presented on 9 February and 25 March 2020 the CHASE project in 'PITCH', a dedicated programme of the Brussels TV channel Bxl1 (https://bx1.be/emission/pitch-nadine-mattielli-ulb/)

8. ENCOUNTERED PROBLEMS AND SOLUTIONS

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

There have been delays in sending back the air cargo from season 2018/2019 which arrived only by beginning of July 2019. This caused delays for the necessary repair of the aerosol in-situ measurement instruments. The aethalometer and the Laser aerosol spectrometer could therefore not sent to PES for season 2019/20.
Also, the air cargo to be sent back from season 2019/20 is currently delayed and will probably only be sent back by sea cargo. The delay is due to staff capacity issues of the station operator in Cape Town and also due to the Covid-19 crisis and corresponding restrictions.

Due to the Covid-19 crisis, participation at (inter-)national scientific conferences is compromised. Many conferences are already cancelled and what will happen with conferences in summer and autumn is not clear yet.

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:

KMI has hired a new staff member who started with 1 December 2019, Dr. Karen De Causmaecker. She will work mainly on atmospheric trajectory calculation and dispersion modelling and contributing thus to analysing the origin of air masses which have been sampled within the Chase project. The employment of Karen De Causmaecker increases the female representation within CHASE.