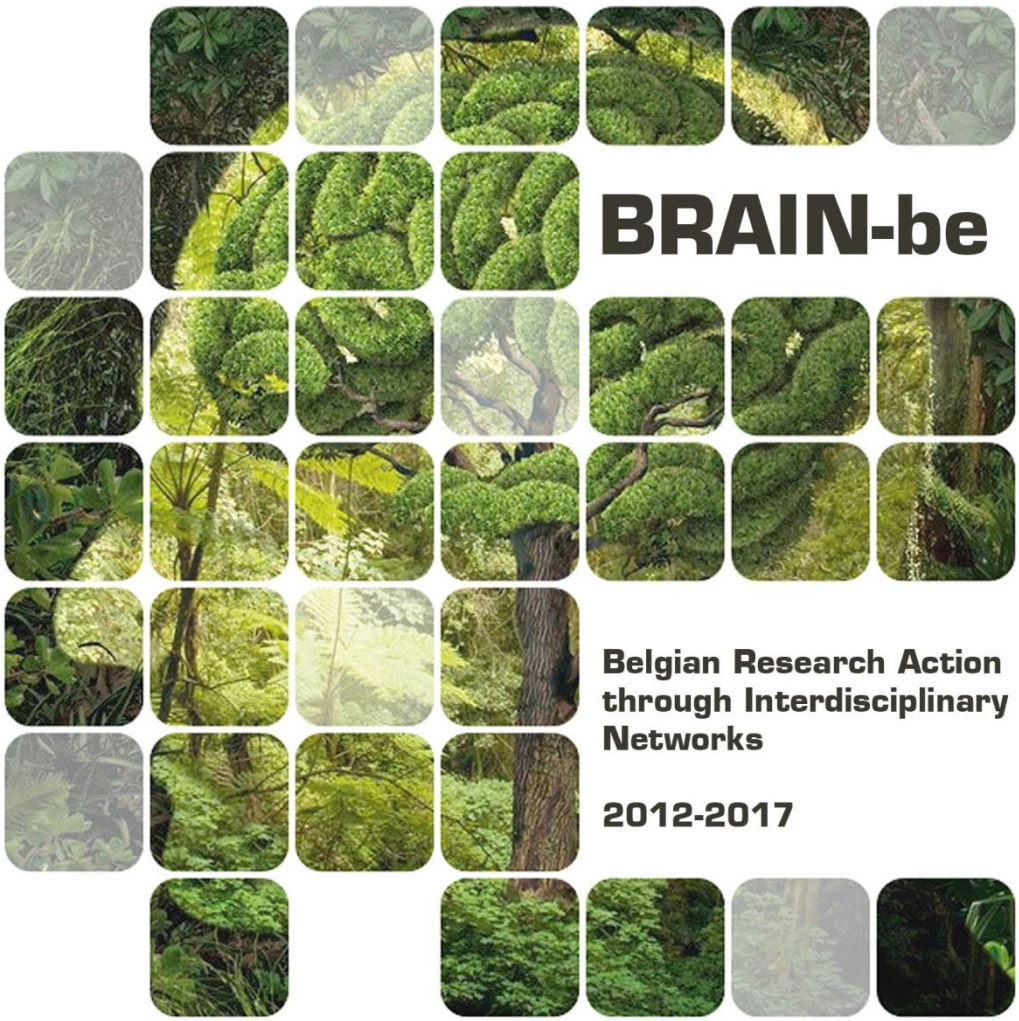

CHASE

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

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NETWORK PROJECT

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Contract - BR/175/A2/CHASE

FINAL REPORT

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ABSTRACT

Context

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

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

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

Objectives

The objectives of CHASE were:

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

Conclusions

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

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

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

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

Keywords

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