

# THE IMPORTANCE OF VILLUM RESEARCH STATION FOR THE DANISH ATMOSPHERIC AMAP CONTRIBUTIONS

Danish contribution to AMAP atmospheric part

Technical Report from DCE - Danish Centre for Environment and Energy No. 165

2020



[Blank page]

# THE IMPORTANCE OF VILLUM RESEARCH STATION FOR THE DANISH ATMOSPHERIC AMAP CONTRIBUTIONS

Danish contribution to AMAP atmospheric part

Technical Report from DCE - Danish Centre for Environment and Energy No. 165

2020

Henrik Skov Rossana Bossi Jesper Christensen Katrin Vorkamp Andreas Massling Jacob Klenø Nøjgaard Kaj Mantzius Hansen Ulas Im Lise Lotte Sørensen

Aarhus University, Department of Environmental Science



# Data sheet

Series title and no.:	Technical Report from DCE - Danish Centre for Environment and Energy No. 165		
Title:	The importance of Villum Research Station for the Danish atmospheric AMAP contributions		
Subtitle:	Danish contribution to AMAP atmospheric part		
Authors:	Henrik Skov, Rossana Bossi, Jesper Christensen, Katrin Vorkamp, Andreas Massling, Jacob Klenø Nøjgaard, Kaj Mantzius Hansen, Ulas Im, Lise Lotte Sørensen		
Institution:	Aarnus University, Department of Environmental Science		
Publisher: URL:	Aarhus University, DCE – Danish Centre for Environment and Energy © http://dce.au.dk/en		
Year of publication: Editing completed:	March 2020 19 <sup>th</sup> February 2020		
Referee: Quality assurance, DCE: Linguistic QA:	Claus Nordstrøm Kirsten Bang Rossana Bossi		
Financial support:	No external financial support		
Please cite as:	Skov, H., Bossi, R., Christensen, J., Vorkamp, K., Massling, A., Nøjgaard, J.K., Kaj Mantzius Hansen, Im, U. & Sørensen, L.L. 2020. The importance of Villum Research Station for the Danish atmospheric AMAP contributions. Danish contribution to AMAP atmospheric part. Aarhus University, DCE – Danish Centre for Environment and Energy, 26 pp. Technical Report No. 165 <u>http://dce2.au.dk/pub/TR165.pdf</u>		
	Reproduction permitted provided the source is explicitly acknowledged		
Abstract:	None		
Keywords:	Atmosphere, monitoring, Arctic Stations		
Layout: Front page photo:	Ann-Katrine Holme Christoffersen Henrik Skov		
ISBN: ISSN (electronic):	978-87-7156-476-1 2245-019X		
Number of pages:	26		
Internet version:	The report is available in electronic format (pdf) at <u>http://dce2.au.dk/pub/TR165.pdf</u>		

# Contents

Pre	eface		5	
Sa	mmer	nfatning (summary in Danish)	6	
Su	mmar	у	7	
1	Intro	oduction	8	
2	Mea	surement sites in Greenland and measured parameters	9	
	2.1	Villum Research Station (AU)	9	
	2.2	Other High Arctic stations in Greenland	10	
	2.3	Other Arctic stations outside Greenland	11	
3	3 Atmospheric models			
	3.1	DEHM model	13	
	3.2	NASA-GISS climate model	14	
	3.3	Statistical models	15	
4	4 The importance of Danish activities in relation to internation			
	oblic	jations and international activities/stations	16	
	4.1	POPs and xenobiotics	16	
	4.2	Heavy metals and plant nutrition salts	18	
	4.3	Mercury	19	
	4.4	Ozone	19	
5	The	strategy of integrated monitoring and outlook	20	
6	Refe	ences	21	

[Blank page]

# Preface

The Arctic Monitoring and Assessment Programme (AMAP) is one of six working groups under the Arctic Council. AMAP has the objective to monitor the levels of anthropogenic pollutants in the Arctic, to document pollutant pathways to and within the Arctic, document the development of pollution and assess the effects of these levels of pollution. Furthermore, AMAP reports to governments and ministers of the Arctic countries and advises on policy matters in relation to Arctic pollution.

AMAP includes representatives of the Arctic countries, permanent participants (i.e. representatives of indigenous people's organization of the Arctic) and a number of observers, which can be countries and organisations. Starting with a first assessment report on Arctic pollution issues in 1998, at the time covering persistent organic pollutants (POPs), heavy metals, radioactivity, acidifying substances, ozone, climate change, human health etc., subsequent assessment reports have focussed on specific fields of pollution or on climate change. The assessment reports address state-of-the-art science and knowledge, compiled and reviewed by scientific expert groups in each field. In addition, indigenous and local knowledge is included.

The assessment reports are delivered to the Arctic Council, and thus to the ministers of the Arctic countries. They usually include science-based policy recommendations. The atmospheric monitoring at Villum Research Station has contributed to several fields assessed by AMAP, including, but not limited to, POPs, heavy metals, short-lived climate forces and climate change. The report is written at the request of The Danish Ministry of Environment and Food and the Energy Agency. DCE/Department of Environmental Science describes in this report the role of Villum Research Station (Villum), at Station Nord in North Greenland in contributing to the knowledge about climate and contaminant loads to the Arctic environment and in particular to Greenland.

# Sammenfatning (summary in Danish)

Arktis er en indikator for globale forandringer og under antropocæn<sup>1</sup> er Arktisk blevet stærkt påvirket af menneskelige aktiviteter. Atmosfæriske teleforbindelser (fx havstrømme som Golfstrømmen og Den Nordatlantiske Strøm, samt vejrsystemer) har tilbagekoblingseffekter på det globale klima og miljø og gør, at forandringer ikke alene er bekymrende for Arktis, men har også indflydelse på global skala. En dobbelt så stor temperaturstigning i Arktis sammenlignet med resten af verden kombineret med øget antropogen aktivitet i Arktis understreger det presserende i at forstå forureningers dynamik og vurdere de deraf afledte konsekvenser.

Formålet med denne rapport er at beskrive betydningen af Villum Research Station (Villum) ved Station Nord i Nordgrønland for vidensopbygningen om forureningsbelastningen til det arktiske miljø og klima, specielt til Grønland. I rapporten beskriver vi, hvordan resultater fra Villum indgår i dokumentationen af atmosfæriske transportveje og dynamikken af forureningsgrupper prioriteret af AMAP.

Vi illustrerer vigtigheden af resultater fra Villum ift. at bestemme hvordan forureningsbelastningen af Arktis påvirkes af ændrede kilder og processer. Ligeledes vises, hvordan kortlevede klimakomponenter interagerer med arktisk klima. Desuden eksemplificeres brugen af måleresultater og resultater fra forskellige typer atmosfæriske modeller. Kombinationen af målinger og modelresultater giver os en evne til at forstå årsagerne til nuværende koncentrationer. Denne viden gør det muligt at forudsige fremtidige niveauer og kilde/recepter-forhold.

# Summary

Arctic is a sentinel for global climate changes. Human activities have a strong impact on the global climate and thus also for the Arctic. Climatic teleconnections (e.g. ocean currents as the Gulf Stream and the North Atlantic Current, as well as weather patterns) gives feedback on the global climate and environment and make the changes relevant not only for the Arctic but for the entire world. A temperature increase in Arctic double as high as the average of the rest of the world together with more human activities highlight the urgency to understand the present dynamics of pollutants in Arctic to assess their effects.

The aim of this report is to describe the role of Villum Research Station (Villum), at Station Nord in North Greenland in contributing to the knowledge about climate and contaminant loads to the Arctic environment and in particular to Greenland. The report will address the importance of Villum for describing the atmospheric transport pathways and dynamics for different contaminant groups and issues prioritised by AMAP.

We will show that the results obtained at Villum are very important for assessing the impact of changing sources and processes controlling the contaminant load to Arctic and how short lived climate forcers interact with climate. Furthermore, we will list examples of the use of measured values at the station and results obtained by different types of atmospheric models. The combination of measurements and model results gives us the ability to understand the causes of present concentration levels. This knowledge makes it possible to predict future levels and source/receptor relationships.

### 1 Introduction

Since the onset of the industrial age starting around 1850, human activities have a more and more visible impact on Earth so that the latest decades are now called Anthropocene<sup>1</sup>. Since 2001, where the term was used for the first time, there are 3,305 hits registered in Web of Science.

One of the consequences of this development is that human induced changes are observed globally including the Arctic. Contaminants emitted at mid-latitudes are long range transported to the Arctic. The  $CO_2$  release from anthropogenic activities leads to global warming, which is more than twice as fast in the Arctic compared to the rest of the world and still accelerating<sup>2</sup>. The lack of political decisions on a global level to reduce  $CO_2$  emission will most likely lead to a temperature increase of 3 degrees at the end of the century and, most likely, we will reach a 1.5-2 degrees temperature increase before 2050. Consequently, abrupt feedback from the environment on climate might occur starting a cascade of effects so e.g. Arctic sea ice melting in Arctic will initiate draught in Amazonia<sup>3-4</sup>.

The Arctic Monitoring and Assessment Program (AMAP) aims to "providing reliable and sufficient information on the status of, and threats to, the Arctic environment, and providing scientific advice on actions to be taken in order to support Arctic governments in their efforts to take remedial and preventive actions relating to contaminants". The Danish Realm contribute to AMAP financed mainly by the means of DANCEA.

The aim of this report is to describe the role of Villum Research Station (Villum), at Station Nord in North Greenland in contributing to the knowledge about climate and contaminant loads to the Arctic environment and in particular to Greenland. The report will address the importance of Villum for describing the atmospheric transport pathways and dynamics for different contaminant groups and issues prioritised by AMAP (selected from AMAP homepage <u>https://arctic-council.org/index.php/en/about-us/working-</u> groups/amap), such as:

- Persistent organic pollutants (POPs)
- Heavy metals (in particular mercury, cadmium, and lead)
- Climate change (environmental consequences and biological effects in the Arctic resulting from global climate change)
- Short lived climate forcers (SLCF)

This report will describe the strategy that we apply in using integrated monitoring<sup>5</sup>, where field measurements are used together with models e.g. the Danish Eulerian Hemispheric Model (DEHM), GISS ModelE-ESM, various statistical receptor.

The created knowledge provides a strong base for the Danish contribution to the Arctic assessment work supporting the Arctic Council, as it will be discussed in this report.

# 2 Measurement sites in Greenland and measured parameters

#### 2.1 Villum Research Station (AU)

Villum Research Station (Villum) (81°36' N, 16°40' W) is located at the military outpost Station Nord in North Greenland. Villum is the only station in Greenland with long-term measurement series of pollutants. The pollutants measured at Villum include heavy metals (e.g. mercury), xenobiotic compounds like polychlorinated biphenyls (PCBs), organochlorine legacy pesticides, brominated flame-retardants, perfluoro alkyl substances (PFAS), ozone, and particles including black carbon (BC). As an important and unique feature, the station is open year-round for visiting scientists. The station can accommodate 14 scientists at a time and in e.g. 2018, there were about 2000 overnight stays. Moreover, the station provides the infrastructure for the Danish contribution to the AMAP atmospheric part.

Continuous atmospheric measurements at Station Nord started in 1990 with a discontinuation between 2002 and 2007, where activities were moved to Lille Malene Mountain uphill Nuuk on the west coast. After a few year at Nuuk, it was decided to move the activities back to Station Nord as long-range transport of pollutants in Nuuk was minor and the concentration levels of pollutants were close to the detection limit of instruments and of chemical physical transport models like DEHM<sup>6-7</sup>. In fact, Station Nord was originally chosen after an intensive campaign from 1979 to 1981 at five locations in Greenland (Thule, Station Nord, Ittoggortoormiit, Qegertarsuag and Ikerasassuaq) where Station Nord was found to be the best location for observing long range transport of pollutants to the Arctic and in particular to the area around Greenland<sup>8-9</sup>. Altogether, we have now 24 years of monitoring data spanning over 30 years. During all these years, the activities have been funded by the Danish EPA by DANCEA means. In 2015, the monitoring program was extended to cover also short-lived Climate Forcers (SLCF) financed by the Danish Energy Agency again by means of DANCEA. The research station has been upgraded twice. The first time was in 1997 by the construction of Flygers Hut financed by DANCEA and then again in 2015, where Villum Research Station was built (named after Villum Kann Rasmussen, the funder of Velux and Villum Foundations) after a grant from the Villum Foundation was obtained. Today Villum stands as one of the leading research facilities for Arctic research. In 2015, four peer-reviewed articles were published with research carried out at Villum, 10 in 2016, 9 in 2017, and 14 in 2018. Until now, we have counted 30 peered reviewed papers in 2019, but more are expected since not all publications are reported back to us immediately after publishing and thus we register them with some delay. Based on the results from Villum we have contributed to several circumpolar AMAP assessment reports as well as one national AMAP report<sup>10</sup>. We are at present participating in three assessment reports on mercury, short-lived climate forcers, contaminants and climate change, respectively.

The peer-reviewed articles are a guarantee of the quality that we provide and give the Danish Realm a strong position in the AMAP assessment work. Today the station has one of the longest time series worldwide of long-range transported atmospheric pollutants from mid latitudes to the Arctic, which makes them more valuable for the assessment work. Villum is host of many projects; below, those from DCE/the Department of Environmental Science – Aarhus University (ENVS-AU) are listed.

Project name	Parameters	Period	Funding source
AMAP CORE,	Atmospheric	Renewed yearly	DANCEA
atmosphere	pollutants <sup>10</sup>		
SLCF project	Short lived	2019 to 1. July	DANCEA
	climate forcers	2020	
Non-target screening	Screening for new	August 2018 to	DANCEA
<ul> <li>– a new approach to</li> </ul>	compounds in 12	September 2020	
identify Arctic pollu-	air samples.		
tants			
ERA-Planet iCUBE	Pollutants in Arctic	2017-2020	EU-H2020
ERA-Planet iGOSP	Hg and POPs	2017-2020	EU-H2020
ICOS	CO <sub>2</sub> and CH <sub>4</sub>	2016-2026	ESFRI
	concentrations		
	and fluxes		
ACTRISSppp and	All atmospheric	2017-2022	EU-H2020
ACTRISSimp	compounds but CO <sub>2</sub>		
	and $CH_4$		
Non-target screening	25 "suspect"	2018-2020	DANCEA
	compounds and un-		
	knowns		
Link phototrophy and	phototrophy and	2020-2022	Villum
methanotrophy in	methanotrophy		Experiments
Glacial Bacteria	in Glaciers		

Table 2.1 Projects carried out by DCE/ENVS at Villum in February 2020.

The core financial support for the atmospheric activities is DANCEA. DANCEA and its predecessor have financed AMAP CORE - atmospheric part since 1990 and have, through the years ensured the financial stability for building up the unique expertise and knowledge that we now have on Arctic atmospheric pollutants where we contribute significantly to the AMAP assessment work. The projects and programs in Table 2.1, as well as past activities, have led to a long series of publications<sup>11-36</sup>. Mostly important, many publications are from international networks, which we are part of, as e.g. AMAP, IASC, IASOA, EMEP, WMO-GAW. Altogether, these publications ensure that AU can deliver highest quality of science-based advice to the political system and to the public. Assessment of trends of pollutants in relation to climate and emission changes requires long term monitoring data. Other Greenlandic stations cannot provide similar long term monitoring data.

#### 2.2 Other High Arctic stations in Greenland

A series of other stations are located in Greenland and on the Greenland ice sheet. A list of all research station on Greenland can be found on <u>https://www.isaaffik.org/</u>. In total, 74 stations are indicated but they vary in size and facilities. The few air pollution monitoring stations in Greenland besides Villum are described below. However, Villum is the only station with long time series of contaminants and systematic selection of parameters related to Arctic Haze.

Summit Station (72° N, 38° W, 3200 m ASL): Details of activities at the station can be found on <u>https://www.geosummit.org/</u>. The first activities date back to 2002. Pollutants originating from biomass burning are measured as well as

a series of greenhouse gasses (e.g. CO<sub>2</sub>, N<sub>2</sub>O, CFCs). Ozone is determined as well. The high altitude of the station makes it ideal for describing pollutants and processes in the free troposphere. Furthermore, the determination of atmospheric components provides an important direct link to understand historical levels observed in ice cores even on a short historical scale<sup>37</sup>.

At Thule airbase the Italian Research Institute CNR has operated particle samplers and the measurements are intensified during the MOSAiC experiment in 2019-2020 (<u>https://mosaic-expedition.org/</u>). The authors could not find information of when those measurements were started. Furthermore, DMI operates a station at Qaanaaq (77.46°N, 69.22°W) that has been operating since the 1950s (<u>https://eu-interact.org/field-sites/the-dmi-geophysical-obser-vatory-qaanaaq/</u>). The station is focusing on meteorological observations.

Zackenberg (74°28' N, 20°34' W) was initially established in 1995 as a biological station but intensive measurements of  $CO_2$  and  $H_2O$  exchange with vegetation have been carried out since 1996 and the first measurements of methane exchange was carried out in 1997 (<u>https://g-e-m.dk/gem-localities/zackenberg/</u>). Besides methane and  $CO_2$ , there is also a climate station that was established already in 1995. The station is a part of the Greenland Ecosystem Monitoring programme.

#### 2.3 Other Arctic stations outside Greenland

A list of circumpolar stations from the International Arctic Systems for observing the Atmosphere (IASOA) is shown in Figure 2.1.

Synoptic data and a detailed description of the stations can be found on the webpage <u>https://www.esrl.noaa.gov/psd/iasoa/science</u> of circumarctic atmospheric stations.

Villum, Zeppelin (Ny Ålesund), Alert, Barrow, Pallas and Tiksi are all atmospheric AMAP stations. A short description of each of the AMAP atmospheric stations outside Greenland is provided below. The results from the stations are all reported to eBAS administered by NILU and can be find on <a href="http://ebas.nilu.no/">http://ebas.nilu.no/</a>.

Zeppelin Research Station at Ny Ålesund (78.9°N, 11.9°E) on Svalbard is located on the eastern side of the Fram strait 610 km east of Villum in the warmer North Atlantic Stream. In the last climate normal (1961-1990) the average yearly temperature at Ny Ålesund was -2°C, but in the latest 10 years the temperature at both stations has increased by more than 2 degrees. Today Ny Ålesund has changed from being a cryospheric station to being a marine station surrounded by open water. It was opened in 1990 with the main aim to look at Arctic haze. Long time series of compounds related to Arctic haze are available (<u>https://www.npolar.no/en/zeppelin/</u>).

Alert (82.5°N,62.3°W) in Canada is located 692 km west of Villum, 200 m above sea level. Alert is influenced by atmospheric transport from the North American continent as well as transport from Eurasia. Alert Background Air Pollution Monitoring Network was started in July 1980. It is thus the oldest high Arctic continued station operated (<u>https://www.canada.ca/en/environment-climate-change/services/science-technology/centres/nunavut.html</u>).



Figure 2.1 Location of IASOA research stations, which also include the AMAP Stations; Villum, Zeppelin (Ny Ålesund, Alert, Barrow and Pallas.

CMDL Barrow ((71.3 $\circ$ N, 156.6 $\circ$ W) is located at the most northern tip of Alaska. It was established in 1970 where measurements of CO<sub>2</sub> were initiated. Since then the programme has been extended several times and is now covering a long suite of compounds and parameters

(https://www.esrl.noaa.gov/gmd/obop/brw/summary.html). Climate related parameters are in focus, and information about the chemical composition of aerosols is limited, whereas there are extensive data on particles and their physical properties and on greenhouse gasses.

Pallas (67.973°N, 24.116°E) is located in North Finland; atmospheric measurements started in 1991. The main research themes include greenhouse gas concentrations and ecosystem–atmosphere fluxes, the climate effects of atmospheric aerosols, aerosol–cloud interactions and air quality. Pallas is one of the sites of the Finnish network for monitoring the concentrations of mercury and other heavy metals, benzo(a)pyrene, ozone and other air pollutants (https://en.ilmatieteenlaitos.fi/pallas-atmosphere-ecosystem-supersite)

There is not any continuously operated AMAP station in north Russia (Siberia, Yamal etc.). Tiksi (71.6°N, 128.9°E) is the station that has had the largest degree of continuity in Russia. Meteorological observations were started in 1932 but most data series started around 2006 and data have been discontinuous since then

(https://www.esrl.noaa.gov/psd/arctic/observatories/tiksi/)

### 3 Atmospheric models

#### 3.1 DEHM model

The Danish Eulerian Hemispheric Model (DEHM) is a chemistry-transport model (CTM), which describes the atmospheric physics and chemistry with a high spatial and temporal resolution. The model domain covers the northern hemisphere and is set-up with a polar stereographic projection centred at the North Pole. DEHM is thus ideally suited for studying Arctic air pollution unlike most other CTMs that applies Cartesian projections and have the Artic area on the border of the domain. The spatial resolution of the grid is 150 km X 150 km with 29 unevenly distributed vertical layers that extend up to 100 hPa with the finest resolution in the atmospheric boundary layer. DEHM is driven by meteorological input from the numerical weather prediction model WRF<sup>38</sup>.

The DEHM model system have been very essential in the international AMAP program and is the only still active atmospheric model which have contributed to many of the assessments in the AMAP program since the first assessment in 1998. In the beginning, DEHM was one of the very few models, which contributes to the AMAP assessment. Today DEHM is one of many atmospheric models but it is still one of the few models, which are dedicated to study the transport into the Arctic, and in that context, in a very close collaboration with the measurement activities at Villum but also with other research stations in the Arctic. The DEHM model has been used in a long series of AMAP reports<sup>31, 39-45</sup>.

The DEHM model was originally developed by the former National Environmental Research Institute (now ATMI - ENVS) to study the atmospheric transport of sulphur and sulphate to the Arctic<sup>46</sup>. The model is continuously developed to include new atmospheric species, new process parameterisations and new features of the model, such as the application of nesting options to study the atmospheric transport with a higher resolution in a limited area of the domain<sup>47</sup>. A full ozone chemistry scheme was implemented to study the transport and deposition of ozone and nitrogen species<sup>48</sup>. Other large developments was the inclusion of lead<sup>49</sup>, CO<sub>2</sub><sup>50</sup> and a mercury chemistry scheme with 7 different Hg species<sup>33, 51</sup>. Surface modules to describe the airsurface exchange processes of POPs, such as α-hexachlorocyclohexane<sup>52-54</sup>, and polychlorinated biphenyls<sup>55-56</sup> has also been implemented. This has also been applied to study new emerging contaminants such as decamethylcyclopentasiloxane (D5)57-59 and Dechlorane plus60. In the recent years, DEHM has also been used to study transport of SLCF's as Black Carbon and Organic Matter in the Arctic and the importance of local sources in Arctic as shipping activities or wildfires.

The combined use of measurements at Villum together with DEHM results provide knowledge about the processes that controls the dynamics of specific pollutants e.g. ozone, Hg, POPs and many others. Based on this new knowledge new parameterisation can be made such as the reactions and deposition of Hg the parameterisation of ozone depletion and the formation of particles in the Arctic atmosphere. With this strategy, we continuously improve the predictability of DEHM.

#### 3.2 NASA-GISS climate model

The GISS Model E-ESM is a newly introduced model at AU and is a fully coupled Earth System Model that describes long-lived compounds in the global environment including atmosphere, sea and terrestrial surfaces. This model is important for the description of long-lived compounds that can be transported across Equator.

NASA Goddard Institute of Space Studies (GISS) Earth System Model (ESM), ModelE2.1, is a fully coupled ESM<sup>61</sup>. A full description of ModelE2.1 and evaluation of its coupled climatology during the satellite era (1979–2014) and the historical ensemble simulation of the atmosphere and ocean component models (1850-2014) are under preparation. ModelE2.1 has a horizontal resolution of 2.0° in latitude by 2.5° in longitude, and 40 vertical layers extending from the surface to 0.1 hPa in the lower mesosphere. The tropospheric chemistry scheme used in ModelE2.1<sup>62-63</sup> includes inorganic chemistry of O<sub>x</sub>, NO<sub>x</sub>, HO<sub>x</sub>, CO, and organic chemistry of CH<sub>4</sub> and higher hydrocarbons using the CBM4 scheme<sup>64</sup>, and the stratospheric chemistry scheme<sup>65</sup>, which includes chlorine and bromine chemistry together with polar stratospheric clouds.

In the AMAP, we are using the OMA, the One-Moment Aerosol scheme<sup>66-71</sup>. OMA is a mass-based scheme in which aerosols are assumed to remain externally mixed and have a prescribed and constant size distribution, with the exception of sea salt that has two distinct size classes, and dust that is described by a sectional model with an option from four to six bins. The scheme treats sulphate, nitrate, ammonium, carbonaceous aerosols (black carbon and organic carbon, including the NO<sub>x</sub>-dependent formation of SOA and methane sulfonic acid formation), dust and sea-salt. The model includes secondary organic aerosol production<sup>72</sup>. The default dust configuration that is used in this work includes five bins, a clay and four silt ones, from submicron to 16 µm in size. The first three dust size bins can be coated by sulphate and nitrate aero-sols<sup>68</sup>. OMA also only includes the first indirect effect. The aerosol number concentration that impacts clouds are obtained from the aerosol mass<sup>73</sup>.

The natural emissions of sea salt, DMS, isoprene and dust are calculated interactively. Anthropogenic dust sources are not represented in ModelE2.1. Dust emissions vary spatially and temporally only with the evolution of climate variables like wind speed and soil moisture<sup>70</sup>.

In frame of the ongoing AMAP assessment for the short-lived climate forcers, we are participating to two sets of experiments. The first experiment concerns historical levels over the Arctic from 1990 to 2015, while the second experiment focuses on fully coupled earth system models, simulating future SLCF levels and climate over the Arctic, employing different emission scenarios. The version of the model we use in the first experiment uses prescribed sea surface temperature (SST) and sea ice fraction during the historical period<sup>74</sup>. The second experiment is particularly important as modelE2 is one of the few models that are participating to this experiment and therefore our contribution is a key element for the assessment of future levels of SLCF and climate in the Arctic.

#### 3.3 Statistical models

In order to develop strategies for controlling and reducing Arctic air pollution, the basic mechanisms for determining the fate of air pollution in the Arctic must be understood. One approach is application of statistical models such as the Constrained Physical Receptor Model (COPREM)75 and Positive Matrix Factorization (PMF) to derive sources of atmospheric particles<sup>29, 76</sup>. In 2013, Nguyen et al. used both receptor models to identify two natural sources (crustal and primary marine factors) and three anthropogenic sources based on 2years of AMAP monitoring data<sup>29</sup>. Although the number of chemical compounds included only elements, ions and elemental carbon in weekly time resolution, the available AMAP data series spans decades and chemical species reflect the sources<sup>49</sup>. On a shorter time scale, PMF has recently been applied to the carbonaceous aerosol only, for quantification of a secondary marine source, primary fossil fuel combustion and a third long-range transported source dominating during Arctic Haze<sup>30</sup>. Both receptor models can be tuned to utilize available data, e.g. short/long time series and few/many chemical species. Thus, receptor models compliment very well Chemical Transport Models like DEHM.

# 4 The importance of Danish activities in relation to international obligations and international activities/stations

Besides being located at one of the most remote locations on Earth, Villum is a high Arctic research station. It is located at the ice stream from the Arctic Ocean that flows down at the westerly part of the Fram Strait, and thus the surroundings are still dominated by ice year round. Villum is thus an ideal location for studying the interaction between the cryosphere and ocean as well as the atmosphere. The other Arctic stations do not have the same opportunities. Furthermore, Villum is located at the east coast of Greenland, where the ice stream enters the Fram Strait from the Arctic Ocean. Multiyear ice and seasonal ice are thus transported right to the door of the station. Ny Ålesund and Zeppelin on Svalbard is located on the eastern side of the Fram strait 610 km east of Villum in the warmer North Atlantic Stream. In the last climate normal (1961-1990) the average yearly temperature at Villum was -16°C compared to -2°C at Ny Ålesund, but in the latest 10 years the temperature at both stations has increased by more than 2 degrees. Today Ny Ålesund has changed from being a cryospheric station to being a marine station surrounded by open water large parts of the year, whereas Villum is still surrounded by sea ice most of the year and there are still years where the ice remains year round.

Alert is located 692 km west of Villum 200 m above sea level. Alert is influenced by atmospheric transport from the North American continent as well as transport from Eurasia, whereas Villum is strongly dominated by atmospheric transport from Eurasia. In a joint article (under preparation), we clearly see that the different stations cover different source regions. Furthermore, processes controlling the pollution levels across Arctic differs and, for example, we have observed different particle formation patterns<sup>77</sup> between the two stations.

Therefore, the results from Villum are very important for assessing the impact of changing sources and processes that control the contaminant load in Arctic and how short lived climate forcers interact with climate.

#### 4.1 POPs and xenobiotics

The UN Stockholm Convention on POPs is a global treaty regulating the production and use of chemicals characterised as POPs (<u>www.pops.int</u>). The screening criteria for a POP classification include long-range transport, persistence, bioaccumulation and adverse effects. Thus, data from the Arctic atmosphere can have an indicator function as they reflect the persistence of a compound and its ability to be transported over long ditances<sup>14, 78</sup>.

Since its adoption in 2001, the Stockholm Convention has regulated 28 POPs according to its annexes A (elimination), B (restriction) or C (unintentional production). The most recent additions include perfluorooctanoic acid (PFOA) and dicofol. In addition, perfluorohexane sulfonate (PFHxS) is currently under review. New candidates that were recently nominated for review by the Stockholm Convention include the insecticide methoxychlor and the chlorinated flame retardant dechlorane plus (DP).

DP was the subject of a recently completed DANCEA project addressing POP and PBT (persistence, bioaccumulation and toxicity) characteristics of DP and other unregulated halogenated flame-retardants and involving air samples from Villum<sup>79</sup>. Initial DP measurements at Villum had been published previously<sup>80</sup>. Results from atmospheric measurements in both studies are included in the proposal document for DP prepared by the POP Review Committee of the Stockholm Convention.

Further data on currently unregulated or rarely studied compounds obtained at Villum were included in the recent AMAP assessments on Chemicals of Emerging Arctic Concern<sup>81</sup>. The contributions to this report have subsequently been published in a special issue of the journal *Emerging Contaminants*. Data from Villum were included in articles on current-use halogenated and organophosphorous flame retardants<sup>82</sup>, poly- and perfluorinated alkylated substances<sup>83</sup> and current-use pesticides<sup>84</sup>.

Besides the indicator function, monitoring data from Villum are used in the effectiveness evaluation of the Stockholm Convention (and potentially other regulatory frameworks for chemicals). Following an international regulation, the concentration of a chemical in the environment is expected to decrease. The POP time series at Villum, comprising a large number of organochlorine, organobromine and organofluorine compounds, document the atmospheric levels and generally show the expected decreases<sup>10, 13-14</sup>. These data are important contributions to AMAP assessments of contaminant time trends<sup>85</sup> and are also included in the contribution from the Western Europe and Other States Group (WEOG), one of five regional coordination groups, to the Global Monitoring Report under the Stockholm Convention. Air is one of three main matrices selected for monitoring under the Stockholm Convention (the others are human blood and human milk as well as water for perfluorooctane sulfonate (PFOS)) where data from Villum have a central role in the global POP assessments.

An ongoing AMAP assessment addresses effects of climate change on contaminants in the Arctic. The four chapters of this assessment report include i) modelling results of emissions and long-range transport of chemicals (in relation to climate change) and ii) contaminant trends in abiotic media (besides changes in food web structures and contaminant trends in biota). In both chapters, modelling results and measurements from Villum are important contributions. For example, measurements at Villum have shown that a reduced ice cover is associated with increased concentrations of  $\alpha$ -HCH in the atmosphere<sup>13</sup>. Increased volatilization from water is one of the expected processes caused by Arctic warming, but few data exist that actually document this.

Members of the AMAP POP expert group recently initiated collaborations on POP modelling, with a view to assist work of the task force on Hemispheric Transport of Air Pollution (HTAP) under the Convention on Long-Range Transboundary Air Pollution (LRTAP). Modelling and monitoring data from Villum are considered in this initiative, through participation of AU in this group, however, it is yet uncertain what specific activities or assessments will be pursued by this group.

#### 4.2 Heavy metals and plant nutrition salts

It is known that during winter and early spring air masses originated at lower mid-latitudes do reach the high Arctic transporting aerosols and gasses emitted in these areas<sup>10</sup>. Some components that have been used and emitted in the industrialized countries in the previous decades were identified in the particulate composition of the Arctic atmosphere and their decline can only be investigated by long-time monitoring activities. Lead (Pb) and arsenic (As) are species typically emitted from combustion sources and do give an indication if various sources are still active. Pb has been banned as an anti-knock agent from gasoline in North America and Europe for many decades and a clear decline has been observed in the Arctic from 1990 and ongoing at Villum<sup>10</sup>. Nevertheless, the emissions from Russia and Asia are not well known and their contribution to the Arctic atmosphere can only be investigated when long-time series of heavy metals are available to carry out source apportionment studies identifying the origin and abundance of these components.

Copper (Cu) und nickel (Ni) are heavy metals, which stem typically from smelters. Such smelters are located in Northern Russia and the transport pathways to the high Arctic are quite efficient<sup>29, 49</sup>. Trend investigations of such components give relevant information about the activity of such smelters, which do introduce environmental risks to the sensible Arctic ecosystems.

In the rising discussion on climate changes, the indirect influence of anthropogenic activities to the climate receives rising attention. In a changing climate with in general higher atmospheric temperatures the melting and retreat of glacier surfaces is a well-known phenomenon that enables soil dust to be reemitted and suspended in air. This process can be understood as a kind of feedback mechanism, in which emissions from e.g. Iceland or Greenland do pollute the Arctic atmosphere with natural components that are observed in Arctic areas. Aluminum (Al) and iron (Fe) are components that represent emissions from soil dust emissions<sup>29</sup> and the continuous monitoring of their atmospheric concentrations over many decades is a relevant activity in order to follow the impact of anthropogenically induced changes in remote environments affecting the Arctic ecosystems. The deposition of soil dust on snow- and ice-covered surfaces actually changes the albedo, which has a direct effect on the radiative balance of the Arctic atmosphere with clear consequences on the climate.

Plant nutrition salts have a special role. Nitrate and sulfate can have both natural sources e.g. sea spray and anthropogenic sources e.g. coal combustion. At Villum, the main source for e.g. sulfate has been identified to come from fossil combustion at mid latitudes<sup>29, 49</sup>.

Villum provides long-term data to AMAP on these salts and metals. Until now, the concentrations in the Arctic of these compounds have been controlled by emission patterns at mid latitudes. In future, this situation might change and climate impact on transport patterns and on processes might be more important. For example, climate has an impact on the strength and position of the Polar Vortex that limits the transport of air pollution from midlatitudes to Arctic during summer. In a warmer climate, the vortex is weakened<sup>86</sup>. We use the data from Villum right now in AMAP assessment work on short-lived climate forcers, where especially sulfate is a strong proxy for anthropogenic particle pollution long transported from mid-latitudes to the Arctic. Furthermore, we also are involved in an assessment report on mercury, see details below.

#### 4.3 Mercury

Mercury in the environment is the first example where human emissions have led to general recommendations of the intake of food from wildlife<sup>42</sup>. The Minemata Convention was signed in Nairobi in 2013, named after the mercury catastrophe in the mid 20ies century in Minemata, Japan. The signing member states agreed to band the manufacturing, use and distribution of mercury (www. mercuryconvention.org). Furthermore, parties agreed to advance a framework to monitor the effectiveness of the convention in order to strengthen its implementation. Villum provide continuous measurements of Gaseous Elemental Mercury (GEM) data and model calculations<sup>10, 33, 51, 87</sup>. In order to improve the understanding of the processes governing the atmospheric Hg concentrations in the Arctic, we have also carried out intensive campaigns where we have studied e.g. the surface exchange and latest the reemission of GEM<sup>88</sup>. Therefore, we are also at the moment, participating in the writing of the new mercury AMAP Assessment Report.

#### 4.4 Ozone

Surface ozone is toxic to humans and noxious towards plants. Besides that, it is also a strong greenhouse gas. Therefore, ozone is also part of various Assessment works under AMAP. At present, we are working on an assessment report on short-lived climate forcers (SLCF) that also include ozone, particle properties and methane.

Ozone is also a key compound for understanding the reactivity of the atmosphere. It is formed when organic compounds are photochemically oxidized in the atmosphere in presence of NOx (NO + NO<sub>2</sub>). Ozone photolysis in the presence of sunlight and water leads to the formation of an OH radical. OH is responsible for the removal of most pollutants, due to its oxidative properties, and it represents the self-cleaning effect of the atmosphere. Ozone measurements are therefore central for understanding the fate of pollutants in the atmosphere<sup>89</sup>. In the Arctic, special reactions occur between ozone and halogen atoms (Cl, Br and I). These special reactions indirectly affects particle formation<sup>90</sup> and also atmospheric mercury<sup>87</sup>. Therefore, our measurement and modelling activities concerning ozone are included in not only the assessment work on SLCF but also in the assessment work on mercury.

## 5 The strategy of integrated monitoring and outlook

The Earth is experiencing accelerating climate changes due mainly to human release of  $CO_2$ . In the Paris Declaration, it was agreed to cut the  $CO_2$  emission so the temperature increase will remain within 1.5 °C compared to preindustrial time. IPCC has estimated that we will reach 1.5 °C already in 2032 to 2050<sup>86</sup> keeping the present climate politics. Because of the current lack of political willingness, we most likely will reach a warming above 2.0 °C at the onset of the next century. Consequently, we cannot predict how climate will impact the ocean, environment, and pollutant transport to and fate in the Arctic. A study of the earth climate system have identified fifteen tipping elements<sup>69</sup>, where the climate will affect the balance of other parameters that in turn will enhance the present radiation balance and thus lead to significant enhanced irreversible temperature increases. Eight of these tipping elements are connected to polar or boreal regions<sup>91</sup>.

Therefore, it is important to study these possible tipping elements. In the last years, we have focused our scientific studies on the role of aerosols in the atmosphere and how aerosols might affect the sea ice extension as well as the melt of glaciers due to their effects on albedo. We anticipate that this work will continue as enhanced cryospheric melting will be a still more imminent problem causing flooding mostly in the southern hemisphere and changes on the major oceanic streams<sup>86</sup> with a global impact.

The warming also leads to changes in human activity patterns in the Arctic and Subarctic areas with more mineral extractions (mining and oil drilling), increased fishing activities and shipping. Altogether, this will change the source regions and types. Furthermore, climate will most likely affect the transport pattern position and strength of the polar front. The polar front is important in regulating the transport of pollutants from mid-latitudes to the Arctic. Therefore, we expect to see changes in the anthropogenic pollution types (dependent on future releases) and source areas.

Villum Research Station at Station Nord is already today an important station for assessing Arctic changes and, due to increased Arctic and global temperature, the Station will be even more important as it is located right in the bull's eye concerning transport of pollutants and where the largest climate change will occur. Measurements at Villum Research Station will provide unique knowledge about the future challenges of emerging pollutants, changes in emission areas, impact of climate on the dynamics of pollutants, transport, atmospheric removal processes as chemical and physical conversion, deposition and reemission as well as storage in e.g. sediments or uptake in biosphere. Combining model and measurement results will in an integrated approach done at Villum, provide indispensable knowledge for policy makers.

# 6 References

- 1. Crutzen, P.J. The "anthropocene". J. Phys. IV 2002, 12 (PR10), 1-5.
- 2. IPCC *IPCC* Fifth assessment report; 2013, 2013.
- 3. Williamson, M.S.; Bathiany, S.; Lenton, T.M., Early warning signals of tipping points in periodically forced systems. *Earth Syst. Dynam.* **2016**, *7* (2), 313-326.
- 4. Lenton, T.M.; Rockstrom, J.; Gaffney, O.; Rahmstorf, S.; Richardson, K.; Steffen, W. and Schellnhuber, H.J. Climate tipping points too risky to bet against. *Nature* **2019**, *575* (7784), 592-595.
- 5. Hertel, O.; Ellermann, T.; Palmgren, F.; Berkowicz, R.; Lofstrom, P.; Frohn, L.M.; Geels, C.; Skjoth, C. A.; Brandt, J.; Christensen, J.; Kemp, K. and Ketzel, M. Integrated air-quality monitoring combined use of measurements and models in monitoring programmes. *Environ. Chem.* **2007**, *4* (2), 65-74.
- 6. Skov, H.; Wahlin, P.; Christensen, J.; Heidam, N.Z. and Petersen, D. Measurements of elements, sulphate and SO2 in Nuuk Greenland. *Atmos. Environ.* **2006**, *40* (25), 4775-4781.
- Bossi, R.; Skov, H.; Vorkamp, K.; Christensen, J.; Rastogi, S.C.; Egelov, A. and Petersen, D. Atmospheric concentrations of organochlorine pesticides, polybrominated diphenyl ethers and polychloronaphthalenes in Nuuk, South-West Greenland. *Atmos. Environ.* 2008, *42* (31), 7293-7303.
- 8. Heidam, N.Z. Crustal Enrichments in the Arctic Aerosol. *Atmos. Environ.* **1985**, *19* (12), 2083-2097.
- 9. Heidam, N.Z., The Components of the Arctic Aerosol. *Atmos. Environ.* **1984**, *18* (2), 329-343.
- Skov, H.; Massling, A.H.; Nielsen, I.E.; Nordstrøm, C.; Bossi, R.; Vorkamp, K.; Christensen, J.; Larsen, M.M.; Hansen, K.M.; Liisberg J.B. and Poulsen M.B. 2017, AMAP CORE - ATMOSPHERIC PART from 1990 to 2015, Results from Villum Research Station. Aarhus University; TR101; Aarhus University: 2017; p 77.
- Angot, H.; Dastoor, A.; De Simone, F.; Gardfeldt, K.; Gencarelli, C.N.; Hedgecock, I.M.; Langer, S.; Magand, O.; Mastromonaco, M.N.; Nordstrom, C.; Pfaffhuber, K.A.; Pirrone, N.; Ryjkov, A.; Selin, N.E.; Skov, H.; Song, S.J.; Sprovieri, F.; Steffen, A.; Toyota, K.; Travnikov, O.; Yang, X. and Dommergue, A. Chemical cycling and deposition of atmospheric mercury in polar regions: review of recent measurements and comparison with models. *Atmos. Chem. Phys.* 2016, *16* (16), 10735-10763.
- 12. Ariya, P.A.; Skov, H.; Grage, M.L. and Goodsite, M.E. Gaseous Elemental Mercury in the Ambient Atmosphere: Review of The Application of Theoretical Calculations and Experimental Studies for Determination of Reaction Coefficients and Mechanisms with Halogens and Other Reactants. *Advances in Quantum Chemistry* **2008**, *55*, *Chapter* 4, 44-54.
- 13. Bossi, R.; Skjoth, C.A. and Skov, H. Three years (2008-2010) of measurements of atmospheric concentrations of organochlorine pesticides (OCPs) at Station Nord, North-East Greenland. *Environmental Science-Processes & Impacts* **2013**, *15* (12), 2213-2219.
- 14. Bossi, R.; Vorkamp, K. and Skov, H. Concentrations of organochlorine pesticides, polybrominated diphenyl ethers and perfluorinated compounds in the atmosphere of North Greenland. *Environ. Pollut.* **2016**, *217*, 4-10.
- Breider, T.J.; Mickley, L.J.; Jacob, D.J.; Ge, C.; Wang, J.; Sulprizio, M.P.; Croft, B.; Ridley, D.A.; McConnell, J.R.; Sharma, S.; Husain, L.; Dutkiewicz, V.A.; Eleftheriadis, K.; Skov, H. and Hopke, P.K. Multidecadal trends in aerosol radiative forcing over the Arctic: Contribution of changes in anthropogenic aerosol to Arctic warming since 1980. *Journal of Geophysical Research-Atmospheres* 2017, *122* (6), 3573-3594.
- Dall'Osto, M.; Geels, C.; Beddows, D. C. S.; Boertmann, D.; Lange, R.; Nojgaard, J. K.; Harrison, R. M.; Simo, R.; Skov, H. and Massling, A. Regions of open water and melting sea ice drive new particle formation in North East Greenland. *Sci Rep* 2018, *8*, 10.
- 17. Douglas, T.A.; Loseto, L.L.; Macdonald, R.W.; Outridge, P.; Dommergue, A.; Poulain, A.; Amyot, M.; Barkay, T.; Berg, T.; Chetelat, J.; Constant, P.; Evans, M.; Ferrari, C.; Gantner, N.; Johnson, M.S.; Kirk, J.; Kroer, N.; Larose, C.; Lean, D.; Muir, D.; Nielsen, T. G.; Poissant, L.; Rognerud, S.; Skov, H.; Sørensen, S.; Wang, F. and Zdanowicz, C.M. The ultimate fate of mercury deposited to arctic marine and terrestrial ecosystems. *Env.Chem.* **2012**.
- Eckhardt, S.; Quennehen, B.; Olivie, D.J.L.; Berntsen, T.K.; Cherian, R.; Christensen, J.H.; Collins, W.; Crepinsek, S.; Daskalakis, N.; Flanner, M.; Herber, A.; Heyes, C.; Hodnebrog, O.; Huang, L.; Kanakidou, M.; Klimont, Z.; Langner, J.; Law, K.S.; Lund, M.T.; Mahmood, R.; Massling, A.; Myriokefalitakis, S.; Nielsen, I.E.; Nojgaard, J.K.; Quaas, J.; Quinn, P.K.; Raut, J.C.; Rumbold, S.T.; Schulz, M.; Sharma, S.; Skeie, R.B.; Skov, H.; Uttal, T.; von Salzen, K. and Stohl, A. Current model capabilities for simulating

black carbon and sulfate concentrations in the Arctic atmosphere: a multi-model evaluation using a comprehensive measurement data set. *Atmos. Chem. Phys.* **2015**, *15* (16), 9413-9433.

- 19. Fenger, M.; Sorensen, L.L.; Kristensen, K.; Jensen, B.; Nguyen, Q.T.; Nojgaard, J.K.; Massling, A.; Skov, H.; Becker, T. and Glasius, M. Sources of anions in aerosols in northeast Greenland during late winter. *Atmos. Chem. Phys.* **2013**, *13* (3), 1569-1578.
- 20. Ferrari, C.P.; Dommergue, A.; Boutron, C.F.; Skov, H.; Goodsite, M. and Jensen, B. Nighttime production of elemental gaseous mercury in interstitial air of snow at Station Nord, Greenland. *Atmos. Environ.* **2004**, *38* (17), 2727-2735.
- 21. Freud, E.; Krejci, R.; Tunved, P.; Leaitch, R.; Nguyen, Q.T.; Massling, A.; Skov, H. and Barrie, L. Pan-Arctic aerosol number size distributions: seasonality and transport patterns. *Atmos. Chem. Phys.* 2017, *17* (13), 8101-8128.
- Hung, H.; Kallenborn, R.; Breivik, K.; Su, Y.S.; Brorstrom-Lunden, E.; Olafsdottir, K.; Thorlacius, J. M.; Leppanen, S.; Bossi, R.; Skov, H.; Mano, S.; Patton, G.W.; Stern, G.; Sverko, E. and Fellin, P. Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993-2006. *Sci. Total Environ.* 2010, *408* (15), 2854-2873.
- 23. Hung, H.; Katsoyiannis, A.A.; Brorstrom-Lunden, E.; Olafsdottir, K.; Aas, W.; Breivik, K.; Bohlin-Nizzetto, P.; Sigurdsson, A.; Hakola, H.; Bossi, R.; Skov, H.; Sverko, E.; Barresi, E.; Fellin, P. and Wilson, S. Temporal trends of Persistent Organic Pollutants (POPs) in arctic air: 20 years of monitoring under the Arctic Monitoring and Assessment Programme (AMAP). *Environ. Pollut.* **2016**, *217*, 52-61.
- 24. Lange, R.; Dall'Osto, M.; Skov, H.; Nojgaard, J.K.; Nielsen, I.E.; Beddowse, D.C.S.; Simob, R.; Harrison, R.M. and Massling, A. Characterization of distinct Arctic aerosol accumulation modes and their sources. *Atmos. Environ.* **2018**, *183*, 1-10.
- 25. Massling, A.; Nielsen, I.E.; Kristensen, D.; Christensen, J.H.; Sorensen, L.L.; Jensen, B.; Nguyen, Q.T.; Nojgaard, J.K.; Glasius, M. and Skov, H. Atmospheric black carbon and sulfate concentrations in Northeast Greenland. *Atmos. Chem. Phys.* **2015**, *15* (16), 9681-9692.
- 26. Moller, A.K.; Barkay, T.; Abu Al-Soud, W.; Sorensen, S.J.; Skov, H. and Kroer, N. Diversity and characterization of mercury-resistant bacteria in snow, freshwater and sea-ice brine from the High Arctic. *FEMS Microbiol. Ecol.* **2011**, *75* (3), 390-401.
- 27. Nguyen, Q.T.; Glasius, M.; Sorensen, L.L.; Jensen, B.; Skov, H.; Birmili, W.; Wiedensohler, A.; Kristensson, A.; Nojgaard, J.K. and Massling, A. Seasonal variation of atmospheric particle number concentrations, new particle formation and atmospheric oxidation capacity at the high Arctic site Villum Research Station, Station Nord. *Atmos. Chem. Phys.* **2016**, *16* (17), 11319-11336.
- 28. Nguyen, Q.T.; Kristensen, T.B.; Hansen, A.M.K.; Skov, H.; Bossi, R.; Massling, A.; Sorensen, L.L.; Bilde, M.; Glasius, M. and Nojgaard, J.K. Characterization of humic-like substances in Arctic aerosols. *Journal of Geophysical Research-Atmospheres* **2014**, *119* (8), 5011-5027.
- 29. Nguyen, Q.T.; Skov, H.; Sorensen, L.L.; Jensen, B.J.; Grube, A.G.; Massling, A.; Glasius, M. and Nojgaard, J.K. Source apportionment of particles at Station Nord, North East Greenland during 2008-2010 using COPREM and PMF analysis. *Atmos. Chem. Phys.* **2013**, *13* (1), 35-49.
- 30. Nielsen, I.E.; Skov, H.; Massling, A.; Eriksson, A.C.; Dall'Osto, M.; Junninen, H.; Sarnela, N.; Lange, R.; Collier, S.; Zhang, Q.; Cappa, C.D. and Nojgaard, J.K. Biogenic and anthropogenic sources of aerosols at the High Arctic site Villum Research Station. *Atmos. Chem. Phys.* **2019**, *19* (15), 18.
- 31. Quinn, P.K.; Stohl, A.; Arneth, A.; Berntsen, T.; Burchart, J.F.; Christensen, J.; Flanner, M.; Kupiainen, K.; Lihavainen, H.; Shepherd, M.; Shevchenko, V.; Skov, H. and Vestreng, V. AMAP, 2011. The Impact of Black Carbon on Arctic Climate; 2011, 2011; p 128 pp.
- 32. Skov, H.; Brooks, S.; Goodsite, M.E.; Lindberg, S.E.; Meyers, T.P.; Landis, M.; Larsen, M.R.B.; Jensen, B.; McConville, G.; Chung, K.H. and Christensen, J. The fluxes of Reactive Gaseous mercury measured with a newly developed method using relaxed eddy accumulation. *Atmos. Environ.* **2006**, *40*, 5452-5463.
- 33. Skov, H.; Christensen, J.H.; Goodsite, M.E.; Heidam, N.Z.; Jensen, B.; Wahlin, P. and Geernaert, G. Fate of elemental mercury in the arctic during atmospheric mercury depletion episodes and the load of atmospheric mercury to the arctic. *Environ. Sci. Technol.* **2004**, *38* (8), 2373-2382.
- Steffen, A.; Douglas, T.; Amyot, M.; Ariya, P.; Aspmo, K.; Berg, T.; Bottenheim, J.; Brooks, S.; Cobbett, F.; Dastoor, A.; Dommergue, A.; Ebinghaus, R.; Ferrari, C.; Gardfeldt, K.; Goodsite, M.E.; Lean, D.; Poulain, A.J.; Scherz, C.; Skov, H.; Sommar, J. and Temme, C. A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow. *Atmos. Chem. Phys.* 2008, *8* (6), 1445-1482.
- 35. Uttal, T.; Starkweather, S.; Drummond, J.R.; Vihma, T.; Makshtas, A.P.; Darby, L.S.; Burkhart, J.F.; Cox, C.J.; Schmeisser, L.N.; Haiden, T.; Maturilli, M.; Shupe, M.D.; De Boer, G.; Saha, A.; Grachev, A.A.; Crepinsek, S.M.; Bruhwiler, L.; Goodison, B.; McArthur, B.; Walden, V.P.; Dlugokencky, E.J.;

Persson, P.O.G.; Lesins, G.; Laurila, T.; Ogren, J.A.; Stone, R.; Long, C.N.; Sharma, S.; Massling, A.; Turner, D.D.; Stanitski, D.M.; Asmi, E.; Aurela, M.; Skov, H.; Eleftheriadis, K.; Virkkula, A.; Platt, A.; Forland, E.J.; Iijima, Y.; Nielsen, I.E.; Bergin, M.H.; Candlish, L.; Zimov, N.S.; Zimov, S.A.; O'Neill, N. T.; Fogal, P.F.; Kivi, R.; Konopleva-Akish, E.A.; Verlinde, J.; Kustov, V.Y.; Vasel, B.; Ivakhov, V.M.; Viisanen, Y. and Intrieri, J.M. International Arctic Systems for Observing the Atmosphere: An International Polar Year Legacy Consortium. *Bull. Amer. Meteorol. Soc.* **2016**, *97* (6), 1033-1056.

- Wex, H.; Huang, L.; Zhang, W.; Hung, H.; Traversi, R.; Becagli, S.; Sheesley, R.J.; Moffett, C.E.; Barrett, T.E.; Bossi, R.; Skov, H.; Hunerbein, A.; Lubitz, J.; Loffler, M.; Linke, O.; Hartmann, M.; Herenz, P. and Stratmann, F. Annual variability of ice-nucleating particle concentrations at different Arctic locations. *Atmos. Chem. Phys.* 2019, *19* (7), 5293-5311.
- 37. Dommergue, A.; Martinerie, P.; Courteaud, J.; Witrant, E. and Etheridge, D.M. A new reconstruction of atmospheric gaseous elemental mercury trend over the last 60 years from Greenland firn records. *Atmos. Environ.* **2016**, *136*, 156-164.
- 38. Skamarock, W.C. and Klemp, J.B. A time-split nonhydrostatic atmospheric model for weather research and forecasting applications. *J. Comput. Phys.* **2008**, *227* (7), 3465-3485.
- 39. AMAP *AMAP Assessment Report: Arctic Pollution Issues.*; Arctic Monitoring and Assessment Programme (AMAP) Oslo, Norway., 1998; p xii+859 pp. .
- 40. AMAP AMAP assessment 2002: Heavy metals in the Arctic; 2005, 2005; p xvi+265 pp.
- 41. AMAP AMAP Assessment 2006: Acidifying Pollutants, Arctic Haze, and Acidification in the Arctic. , ; Arctic Monitoring and Assessment Programme (AMAP): Oslo, Norway., 2006; p xii+112 pp.
- 42. AMAP AMAP Assessment: Mercury in the Arctic.; Oslo, Norway, 2011, 2011; p xiv + 193 pp.
- 43. Kallenborn, K.; Borga, K.; Christensen, J.H.; Dowdall, M.; Evenset, A.; Odland, J.Ø.; Ruus, A.; Aspmo Pfaffhuber, K.; Pawlak, J. and Reiersen L.-O. *Combined Effects of Selected Pollutants and Climate Change in the Arctic Environment*; Oslo, Norway, 2011; p 108 pp.
- Carlsson, P.; Christensen, J.H.; Borga, K.; Kallenborn, R.; Aspmo Pfaffhuber, K.; Odland, J.Ø.; Reiersen, L.-O. and Pawlak, J.F. *Influence of Climate Change on Transport, Levels, and Effects of Contaminants in Northern Areas – Part 2*; Oslo, Norway, 2017; p 52 pp.
- 45. AMAP, AMAP Assessment 2015: Black carbon and ozone as Arctic climate forcers. *Arctic Monitoring and Assessment programme (AMAP)* 2015, (*WWW.AMAP.NO*) 116pp.
- 46. Christensen, J.H. The Danish Eulerian hemispheric model A three-dimensional air pollution model used for the Arctic. *Atmos. Environ.* **1997**, *31* (24), 4169-4191.
- 47. Frohn, L.M.; Christensen, J.H. and Brandt, J. Development of a high-resolution nested air pollution model The numerical approach. *J. Comput. Phys.* **2002**, *179* (1), 68-94.
- 48. Brandt, J.; Silver, J.D.; Frohn, L.M.; Geels, C.; Gross, A.; Hansen, A.B.; Hansen, K.M.; Hedegaard, G. B.; Skjoth, C.A.; Villadsen, H.; Zare, A. and Christensen, J.H. An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport of air pollution. *Atmos. Environ.* **2012**, *53*, 156-176.
- 49. Heidam, N.Z.; Christensen, J.; Wahlin, P. and Skov, H. Arctic atmospheric contaminants in NE Greenland: levels, variations, origins, transport, transformations and trends 1990-2001. *Sci. Total Environ.* **2004**, *331* (1-3), 5-28.
- 50. Geels, C.; Gloor, M.; Ciais, P.; Bousquet, P.; Peylin, P.; Vermeulen, A.T.; Dargaville, R.; Aalto, T.; Brandt, J.; Christensen, J.H.; Frohn, L.M.; Haszpra, L.; Karstens, U.; Rodenbeck, C.; Ramonet, M.; Carboni, G. and Santaguida, R. Comparing atmospheric transport models for future regional inversions over Europe - Part 1: mapping the atmospheric CO2 signals. *Atmos. Chem. Phys.* **2007**, 7 (13), 3461-3479.
- 51. Christensen, J.H.; Brandt, J.; Frohn, L.M. and Skov, H. Modelling of mercury in the Arctic with the Danish Eulerian Hemispheric Model. *Atmos. Chem. Phys.* **2004**, *4*, 2251-2257.
- 52. Hansen, K.M.; Christensen, J.H.; Brandt, J.; Frohn, L.M.; Geels, C. Modelling atmospheric transport of alpha-hexachlorocyclohexane in the Northern Hemisphere with a 3-D dynamical model: DEHM-POP. *Atmos. Chem. Phys.* **2004**, *4*, 1125-1137.
- 53. Hansen, K.M.; Halsall, C. J.; Christensen, J.H.; Brandt, J.; Frohn, L.M.; Geels, C. and Skjoth, C.A. The role of the snowpack on the fate of alpha-HCH in an atmospheric chemistry-transport model. *Environ. Sci. Technol.* **2008**, *42* (8), 2943-2948.
- 54. Hansen, K.M.; Halsall, C.J. and Christensen, J.H. A dynamic model to study the exchange of gasphase persistent organic pollutants between air and a seasonal snowpack (vol 40, pg 2644, 2006). *Environ. Sci. Technol.* **2008**, *42* (6), 2205-2206.

- 55. Hansen, K.M.; Christensen, J.H.; Geels, C.; Silver, J.D. and Brandt, J. Modelling the impact of climate change on the atmospheric transport and the fate of persistent organic pollutants in the Arctic. *Atmos. Chem. Phys.* **2015**, *15* (11), 6549-6559.
- 56. Carlsson, P.; Breivik, K.; Brorstrom-Lunden, E.; Cousins, I.; Christensen, J.; Grimalt, J.O.; Halsall, C.; Kallenborn, R.; Abass, K.; Lammel, G.; Munthe, J.; MacLeod, M.; Odland, J.Ø.; Pawlak, J.; Rautio, A.; Reiersen, L.O.; Schlabach, M.; Stemmler, I.; Wilson, S. and Wohrnschimmel, H. Polychlorinated biphenyls (PCBs) as sentinels for the elucidation of Arctic environmental change processes: a comprehensive review combined with ArcRisk project results. *Environmental Science and Pollution Research* 2018, *25* (23), 22499-22528.
- 57. McLachlan, M.S.; Kierkegaard, A.; Hansen, K.M.; van Egmond, R.; Christensen, J.H. and Skjoth, C.A. Concentrations and Fate of Decamethylcyclopentasiloxane (D-5) in the Atmosphere. *Environ. Sci. Technol.* **2010**, *44* (14), 5365-5370.
- 58. Genualdi, S.; Harner, T.; Cheng, Y.; MacLeod, M.; Hansen, K.M.; van Egmond, R.; Shoeib, M. and Lee, S. C. Global Distribution of Linear and Cyclic Volatile Methyl Siloxanes in Air. *Environ. Sci. Technol.* **2011**, *45* (8), 3349-3354.
- 59. Krogseth, I.S.; Kierkegaard, A.; McLachlan, M.S.; Breivik, K.; Hansen, K.M. and Schlabach, M. Occurrence and Seasonality of Cyclic Volatile Methyl Siloxanes in Arctic Air. *Environ. Sci. Technol.* **2013**, *47* (1), 502-509.
- 60. Hansen, K. e. a., Global Emissions of Dechlorane Plus. Sci. Total Environ. 2020, In prep. .
- 61. Schmidt, G.A.; Kelley, M.; Nazarenko, L.; Ruedy, R.; Russell, G.L.; Aleinov, I.; Bauer, M.; Bauer, S.E.; Bhat, M.K.; Bleck, R.; Canuto, V.; Chen, Y.H.; Cheng, Y.; Clune, T.L.; Del Genio, A.; de Fainchtein, R.; Faluvegi, G.; Hansen, J.E.; Healy, R.J.; Kiang, N.Y.; Koch, D.; Lacis, A.A.; LeGrande, A.N.; Lerner, J.; Lo, K.K.; Matthews, E.E.; Menon, S.; Miller, R.L.; Oinas, V.; Oloso, A.O.; Perlwitz, J.P.; Puma, M.J.; Putman, W.M.; Rind, D.; Romanou, A.; Sato, M.; Shindell, D.T.; Sun, S.; Syed, R.A.; Tausnev, N.; Tsigaridis, K.; Unger, N.; Voulgarakis, A.; Yao, M.S. and Zhang, J.L. Configuration and assessment of the GISS ModelE2 contributions to the CMIP5 archive. *Journal of Advances in Modeling Earth Systems* 2014, 6 (1), 141-184.
- 62. Shindell, D.T.; Grenfell, J.L.; Rind, D.; Grewe, V. and Price, C. Chemistry-climate interactions in the Goddard Institute for Space Studies general circulation model 1. Tropospheric chemistry model description and evaluation. *Journal of Geophysical Research-Atmospheres* **2001**, *106* (D8), 8047-8075.
- 63. Shindell, D.T.; Faluvegi, G. and Bell, N., Preindustrial-to-present-day radiative forcing by tropospheric ozone from improved simulations with the GISS chemistry-climate GCM. *Atmos. Chem. Phys.* **2003**, *3*, 1675-1702.
- 64. Gery, M.W.; Whitten, G.Z.; Killus, J.P.; Dodge, M.C. A PHOTOCHEMICAL KINETICS MECHANISM FOR URBAN AND REGIONAL SCALE COMPUTER MODELING. Journal of Geophysical Research-Atmospheres **1989**, *94* (D10), 12925-12956.
- 65. Shindell, D.T.; Faluvegi, G.; Unger, N.; Aguilar, E.; Schmidt, G.A.; Koch, D.M.; Bauer, S.E. and Miller, R.L. Simulations of preindustrial, present-day, and 2100 conditions in the NASA GISS composition and climate model G-PUCCINI. *Atmos. Chem. Phys.* **2006**, *6*, 4427-4459.
- 66. Bauer, S.E.; Koch, D.; Unger, N.; Metzger, S.M.; Shindell, D.T.; Streets, D.G., Nitrate aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone. *Atmos. Chem. Phys.* 2007, 7 (19), 5043-5059.
- 67. Bauer, S.E.; Mishchenko, M.I.; Lacis, A.A.; Zhang, S.; Perlwitz, J. and Metzger, S.M. Do sulfate and nitrate coatings on mineral dust have important effects on radiative properties and climate modeling? *Journal of Geophysical Research-Atmospheres* **2007**, *112* (D6), 9.
- 68. Bauer, S.E.; Koch, D. Impact of heterogeneous sulfate formation at mineral dust surfaces on aerosol loads and radiative forcing in the Goddard Institute for Space Studies general circulation model. *Journal of Geophysical Research-Atmospheres* **2005**, *110* (D17), 15.
- 69. Koch, D.; Schmidt, G.A.; Field, C.V. Sulfur, sea salt, and radionuclide aerosols in GISS ModelE. *Journal of Geophysical Research-Atmospheres* **2006**, *111* (D6), 26.
- 70. Miller, R.L.; Cakmur, R.V.; Perlwitz, J.; Geogdzhayev, I.V.; Ginoux, P.; Koch, D.; Kohfeld, K.E.; Prigent, C.; Ruedy, R.; Schmidt, G.A.; Tegen, I. Mineral dust aerosols in the NASA goddard institute for Space Sciences ModelE atmospheric general circulation model. *Journal of Geophysical Research-Atmospheres* **2006**, *111* (D6), 19.
- 71. Tsigaridis, K.; Koch, D. and Menon, S. Uncertainties and importance of sea spray composition on aerosol direct and indirect effects. *Journal of Geophysical Research-Atmospheres* **2013**, *118* (1), 220-235.
- 72. Tsigaridis, K. and Kanakidou, M. Secondary organic aerosol importance in the future atmosphere. *Atmos. Environ.* **2007**, *41* (22), 4682-4692.

- 73. Menon, S. and Rotstayn, L. The radiative influence of aerosol effects on liquid-phase cumulus and stratiform clouds based on sensitivity studies with two climate models. *Clim. Dyn.* **2006**, *27* (4), 345-356.
- 74. Rayner, N.A.; Parker, D.E.; Horton, E.B.; Folland, C.K.; Alexander, L.V.; Rowell, D.P.; Kent, E.C. and Kaplan, A. Global analyses of sea surface temperature, sea ice, and night marine air temperature since the late nineteenth century. *Journal of Geophysical Research-Atmospheres* **2003**, *108* (D14), 37.
- 75. Wählin, P. COPREM A multivariate receptor model with a physical approach. *Atmos. Environ.* **2003**, *37* (35), 4861-4867.
- 76. Skov, H.; Bossi, R.; Wahlin, P.; Vikelsøe, J.; Christensen, J.; Egeløv, A.H.; Heidam, N.Z.; Jensen, B.; Ahleson, H.P.; Stausgard, L.; Jensen, I. and Petersen, D. *Contaminants in the Atmosphere Subtitle: AMAP- Nuuk, Westgreenland 2002-2004*; 547; 2005, 2005.
- 77. Beck, L.S.N.; Junninen, H.; Hoppe, C.J.M.; Garmash, O.; Bianchi, F.; Riva, M.; Rose, C.; Peräkylä, O.; Wimmer, D.; Kausiala, O.; Jokinen, T.; Ahonen, L.; Mikkilä, J.; Hakala, J.; Wolf, K.K.E.; Cappelletti, D.; Mazzola, M.; Traversi, R.; Petroselli, C.; Viola, A.P.; Vitale, V. Lange, R.; Massling, A.; Nøjgaard, J.K.; Krejci, R.; Karlsson, L.; Ziegler, P.; Jang, S.M; Lee, K.; Vakkari, V.; Lampilahti, J.; Thakur, R.C.; Leino, K.; Kangasluoma, J.; Duplissy, E.-M.; Siivola, E.; Kontkanen, J.; Marbouti, M.; He, X.-C.; Tham, Y.J.; Saiz-Lopez, A.; Petäjä, T.; Ehn, M.; Worsnop, D.R.; Skov, H.; Kulmala, M.; Kerminen, V.-M. and Sipilä, M. Aerosol formation is closely linked to changes in the Arctic ecosystem. *Science* 2020, *Submitted January 2020*.
- 78. Vorkamp, K. and Riget, F. Kemikalier i et globalt perspektiv. *Miljøog Sundhed* **2019**, *25* (2), 22-27.
- 79. Vorkamp, K.; Riget, F.; Sanderson, H.; Bossi, R., Hansen, K.M. and Skov, H. *POP/PBT characterisation of dechlorane plus and novel brominated flame retardants on the basis of data from Greenland*; Aarhus University: 2019; SR339, p 80.
- 80. Vorkamp, K.; Bossi, R.; Riget, F.; Skov, H.; Sonne, C. and Dietz, R. Novel brominated flame retardants and dechlorane plus in Greenland air and biota. *Environ. Pollut.* **2015**, *196*, 284-291.
- 81. AMAP Chemicals of Emerging Arctic Concern. Arctic Monitoring and Assessment Programme Oslo Norway, 2017; p XVI + 353.
- 82. Vorkamp, K.; Balmer, J.; Hung, H.; Letcher, R.J.; Rigét, F.F. and de Wit, C.A. Current-use halogenated and organophosphorous flame retardants: A review of their presence in Arctic ecosystems. *Emerging Contaminants* **2019**, *5*, 179-200.
- Muir, D.; Bossi. R.; Carlsson, P.; Evans, M.; De Silva, A.; Halsall, C.; Rauert, C.; Herzke, D.; Hung, H.; Letcher, R.; Rigét, F. and Roos, A. Levels and trends of poly- and perfluoroalkyl substances in the Arctic environment An update. *Emerging Contaminants* 2019, *5*, 240-271.
- 84. Balmer, J.E.; Morris, A.D.; Hung, H.; Jantunen, L.; Vorkamp, K.; Rigét, F.; Evans, M.; Houde, M. and Muir, D.C.G., Levels and trends of current-use pesticides (CUPs) in the Arctic: An updated review. *Emerging Contaminants* **2019**, *5* (70-88).
- 85. AMAP *Temporal trends in persistent organic pollutants in the Arctic*; Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, 2016; p vi + 71.
- 86. IPCC Global warming of 1.5°C; www.ipcc.ch, October 2018, 2018.
- 87. Skov, H.; H. J. N., C.; Jensen, B.; Christoffersen, C.; Poulsen, M.B.; Liisberg, J.B. and Dall'Osto, M., The variability of ozone and mercury at Villum Research Station, Station Nord in North Greenland from 1995 to 2017. *Submitted to ACPD* **2019-October**.
- 88. Kamp, J.; Skov, H.; Jensen, B.; Sorensen, L.L. Fluxes of gaseous elemental mercury (GEM) in the High Arctic during atmospheric mercury depletion events (AMDEs). *Atmos. Chem. Phys.* **2018**, *18* (9), 6923-6938.
- 89. Seinfeld, J.H.; Pandis, S.N., *Atmospheric chemistry and physics: from Air Pollution to climate change*. Second ed.; Wiley-Interscience: 2006.
- 90. Sipila, M.; Sarnela, N.; Jokinen, T.; Henschel, H.; Junninen, H.; Kontkanen, J.; Richters, S.; Kangasluoma, J.; Franchin, A.; Perakyla, O.; Rissanen, M.P.; Ehn, M.; Vehkamaki, H.; Kurten, T.; Berndt, T.; Petaja, T.; Worsnop, D.; Ceburnis, D.; Kerminen, V.M.; Kulmala, M.; O'Dowd, C. Molecular-scale evidence of aerosol particle formation via sequential addition of HIO3. *Nature* 2016, 537 (7621), 532-534.
- 91. Lenton, T. M.; Held, H.; Kriegler, E.; Hall, J.W.; Lucht, W.; Rahmstorf, S. and Schellnhuber, H.J., Tipping elements in the Earth's climate system. *Proc. Natl. Acad. Sci. U. S. A.* **2008**, *105* (6), 1786-1793.Sammenfatning.

[Blank page]

#### THE IMPORTANCE OF VILLUM RESEARCH STATION FOR THE DANISH ATMOSPHERIC AMAP CONTRIBUTIONS

Danish contribution to AMAP atmospheric part

ISBN: 978-87-7156-476-1 ISSN: 2245-019X